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STUDIES ON THE RADIOACTIVE CONTAMINATION DUE TO NUCLEAR DETONATIONS I-VI

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(Studies on the Radioactive Dust due to Nuclear Detonation in Bikini on March 1, 1954)

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Studies on the Radioactive Dust due to Nuclear Detonation in Bikini on March 1, 1954.

An unusually large amount of strong radioactive ash was produced by the thermonuclear test conducted on March 1, 1954 at the Bikini Atoll in the South Pacific by the United States Atomic Energy Commission.

A Japanese fishing boat celled No.5 Fukuryu Maru which was engaged in fishing about 80 - 90 miles east of Bikini early in the morning of March 1 was showered by this ash. The boat, contaminated by the ash came back to Japan in the middle of March with the crew apparently injured by the strong radiation emitted from the ash. The contaminated fish brought back by this boat had been sent to various parts of Japan including Osaka and some of them had been distributed in the market before radiation monitoring.

Under such circumstances, to meet the urgent needs of public health, the studies on the radioactivity of Bikini ash and the radioactive contamination of environment have been started, from the health physics standpoint, with the initiative of the author under close cooperation with the public health officers of local governments in Osaka district since the middle of March, 1954, when the author was the head of the Department of Biopysics, Osaka City University, School of Medicine.

The estimation of the probable dose of radiation the crew might have received during their voyage and the accurate estimation of beta-ray energies and the detection of alpha-ray activity as well as the identification of various radioactive nuclides included in the Bikini ash were considered to be urgently needed items of information in estimating the possible hazard due to the internal

as well as the external irradiation from the health physics point of view.

1. Radioactive Dust

On the morning of March 16, 1954, the author was requested by the public health authorities of Osaka to go to the Osaka City Central Market to examine some of the tuna fish which had been brought back by the No.5 Fukuryu Maru and sent down to Osaka.

On rough examination of the fish at the market it was found that most of them were emitting about 2,000 cpm on the average as measured a few centimeters distant from the wet skin with a Geiger-Muller counter with 1.9 mg/cm^2 mica window and effective area about π cm^2. The natural count of this beta-ray counter was about 20 cpm. For the purpose of comparison some other fish which had been caught near the coast of Japan were also examined, but no significant difference from the natural count could be detected.

On careful examination in the laboratory on March 16, some of the most contaminated fish were found to be emitting over 10,000 cpm per cm^2 of the skin, which may be estimated to correspond roughly to about 0.1 - 1 micro curie per cm^2. The average contamination was probably about one tenth of this value. Dried shark fins were also found to be strongly contaminated. Although the fish caught later were found to be contaminated mainly in the internal organs, those fish brought back by the No.5 Fukuryu Maru were found to be more contaminated on the skin.

The absorption curve of the radiation emitted from the surface of the contaminated tuna fish obtained at Osaka City Central Market on March 16 is shown in Fig.1, and the decay curve of gross activity in Fig.2.

(Fig.1)
(Fig.2)

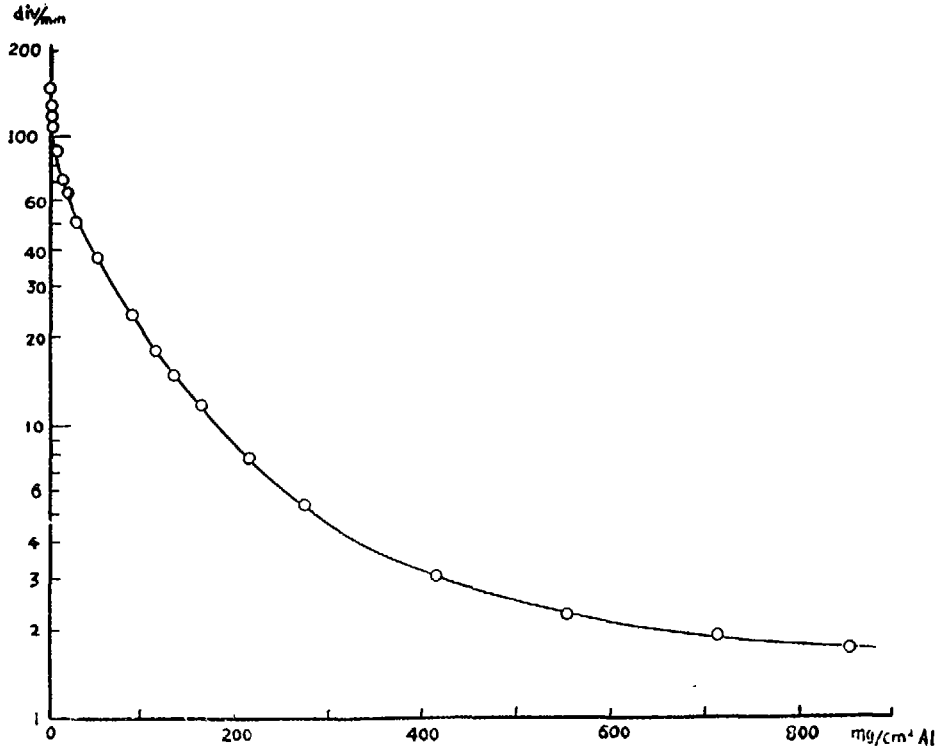

Fig. 1. The absorption curve of the radiation emitted from the surface of tuna fish on March 26, 1954. (Measurement was made at 2 cm from the 3.5 mg/cm² aluminium foil window of a Lauritsen-type electrometer manufactured by Scientific Research Institute, Ltd, Tokyo, Japan.)

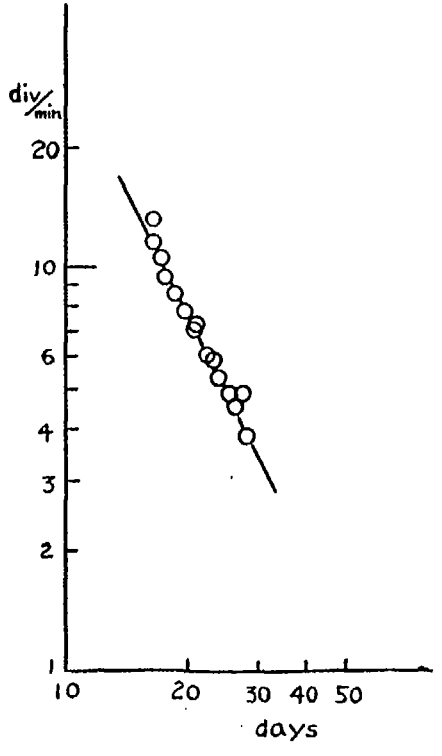

Fig. 2. The decay curve of radioactivity detected from the surface of tuna fish obtained at the Osaka City Central Market on March 16, 1954. (Lauritsen-type electrometer with 3.5 mg/cm^2 Al window)
$A = A_1 t^{-2.05 \pm 0.05}$, $16d < t < 30d$
(t : No. of days after explosion)

Because of the unexpectedly strong radioactivity detected on the contaminated fish in Osaka, it was felt to be very important to go down to the fishing port, Yaizu, immediately to examine the radioactivity of the No.5 Fukuryu Maru, the boat, on which the contaminated fish had been brought back to Japan. On the morning of March 17, the author arrived at Yaizu and collected some radioactive dust from the upper deck of the steering room of the boat with his staff members of the Department of Biophysics, Osaka City University. The lower deck seemed to be thoroughly washed, and yet strong radiation was being emitted. The radioactive contamination of some of the samples brought back from the boat is shown in Table 1.

(Table 1)

On that morning of March 17, the readings of a portable, ionization chamber indicated that the radiation dose rate then being received on the boat was about 80 - 120 mr/h. Extrapolating back ($t^{-1.4}$) from these figures, the possible dose of external radiation which some of the crew might have been receiving during the initial 24 hours period can be estimated roughly at about 200 300 r, and that during the 13 days voyage (March 1 - 14) at about 300 - 500 r. Since this does not take into account the radioactive ash which was washed away by the crew during the voyage, the initial dose must have been considerably greater.

From later examination it was found that tiny white or greyish white particles of diameter about 0.1 mm or less were emitting strong radiation. On microscopic examination even smaller particles, the size of a few microns, were also found. Occasionally, similar particles of a larger size with less radioactivity were observed, but on careful examination these seemed to be the pulverized white paint of the boat which may have become contaminated later by the radioactive dust. The absorption curve and the decay curve of gross activity of the radioactive dust (Bikini ash) are shown in Figs.3 and 4 respectively.

Table 1. Radioactive contamination in cpm of some of the samples collected from the No. 5 Fukuryu Maru on the morning of March 17, 1954. Measurement was made during the period March 18~20, 1954, at about 3 cm from the mica window (1.9 mg/cm^2) of the beta-ray counter whose natural count is about 20 cpm.

Item	Weight in grams	Cpm above natural
Rope	0.2	7047
Cigarette box	0.6	173
Scraped piece of the washed wooden board of the lower deck	0.4	1828
Wood dust	0.3	37307
Tip of the brush	0.3	44312
Cotton glove	0.5	14222
Chop stick	0.2	287
Cabbage	0.6	205
Tobacco	0.2	710
Radish	1.2	314
Scraped piece of the washed wooden part at the head of the boat	0.3	2348
Iron dust (rust)	1.7	11577
Washed rain coat (shoulder part)	0.7	508
" " " (chest part)	0.5	123
Button of the coat	1.0	1275
Cigarette butts	0.5	199
Soap	1.2	325
Canvas	0.2	1349
Small rope (inside)	0.7	4592
Small rope (on the deck)	0.5	15167
Dead cockroaches (from kitchen)	0.5	2788
Rag in the cabin	0.2	4622
Water in the cabin	0.5	387
Hair	0.2	6030
Cover of the canned food	8 g	18135
Iron cable	121 g	66540

(Fig.3)
(Fig.4)

Some of the most radioactive particles, with a size of 0.1 - 0.3 mm, were estimated to have radioactivity of about 0.42 - 1.53 mc/g on March 21. Extrapolating back, it was estimated that one gram of this dust might have had radioactivity equivalent to about 0.5 - 1.5 curies at the time it fell on the boat (7 - 11:30 a.m. March 1). The radioactivity per unit volume was occasionally found to be more than ten times stronger with the particles of smaller sizes (a few to a few tens of microns). One gram of fission products at about three hours after nuclear fission may be estimated to have an activity of about three million curies, assuming the following relation for the gross beta activity,

$$A_\beta = 12 \times 10^6 \, t^{-1.2} \quad \text{curies/gram} \qquad (1)$$

where t is the time after nuclear fission expressed in unit of hour.

Therefore we may assume that a dilution by a factor of about three million might have occurred at the time of detonation.

On preliminary radiochemical analysis conducted on March 18, about 50 - 60% of the apparent beta-ray activity was found to be deposited in the fraction of rare-earth elements mixture with lanthanum as a carrier, although the main constituent of the ash itself seemed to be a calcium compound. This result was consistent with the results obtained by a more detailed radiochemical analysis conducted later as well as with those obtained by others.[1)-(5),(19)(20)]

2. Radioactive Contamination of the Fishermen

While in Yaizu on March 17, the author had the opportunity of examining some of the crew of the No.5 Fukuryu Maru through the courtesy of Dr. Ooi of Kyoritsu Yaizu Hospital. Examination

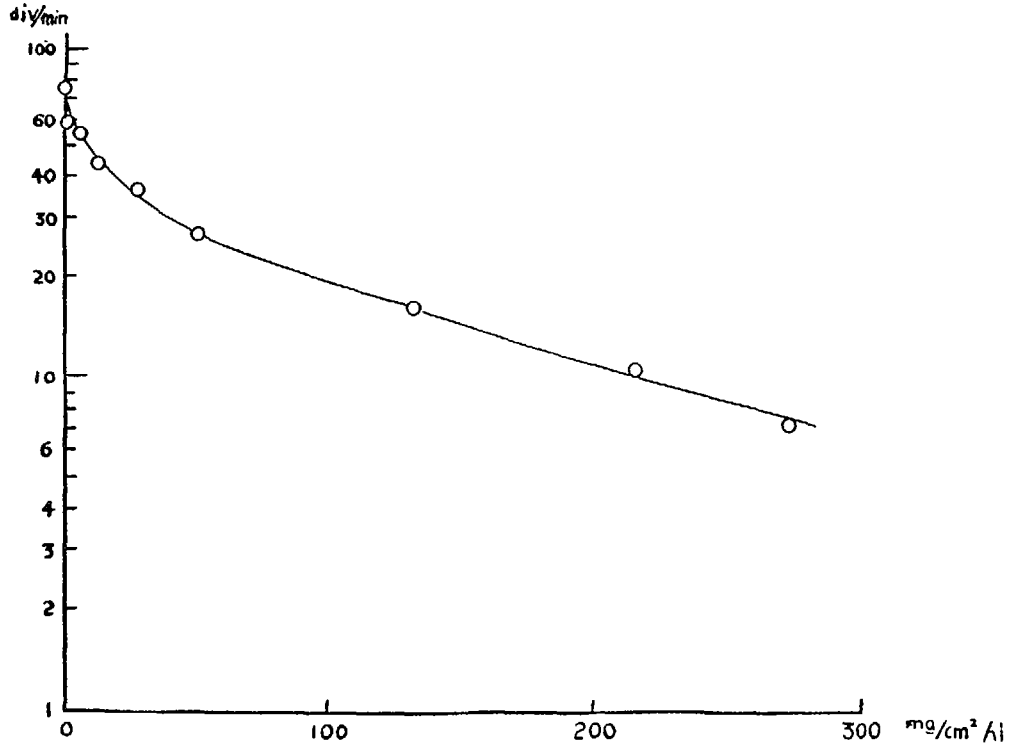

Fig. 3. The absorption curve of the radioactive dust (Bikini ash) collected from the No. 5 Fukuryu Maru. (Lauritsentype electrometer with 3.5 mg/cm² Al window).

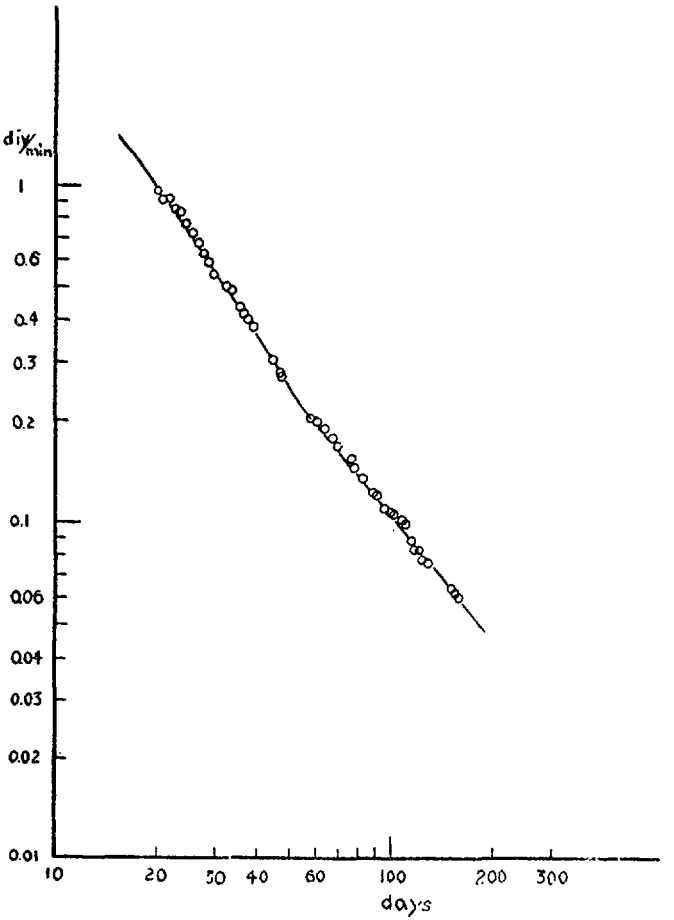

Fig. 4. The decay curve of Bikini ash (greyish white) in logarithmic plot. (Lauritsen-type electrometer with 3.5 mg/cm² Al window)

$A = A_1 t^{-1.59 \pm 0.02}$, $20d \leq t \leq 60d$.

$A = A'_1 t^{-1.22 \pm 0.01}$, $60d \leq t \leq 170d$.

$A = A''_1 t^{-1.38 \pm 0.05}$, $20d \leq t \leq 170d$.

(t : No. of days after explosion)

of the patients in the hospital clearly revealed that radiation was being emitted from the hair, face, neck, and nails. Radioactivity above the naturally occurring level was distinctly detected in the blood, urine, and feces during those days as shown in Table 2. The fishermen's lavatory was also found to

(Table 2)

be slightly radioactive. According to statements by some of the crew, the whitish dust fell so heavily that they could hardly keep their eyes and mouths open.

The condition on the boat, the No.5 Fukuryu Maru, at the time it was being showered by the radioactive ash on March 1, 1954, was experimentarily reproduced by Dr.Miyoshi of Tokyo University Hospital(6) by using pulverized coral reef. This experiment was carried out in the presence of the crew as witnesses to the actual amount of ash which had fallen on the boat. This amount was then estimated to be approximately 3.83 -8.52 mg/cm^2. Judging from this figure, the amount of radioactivity per square meter on the boat can be estimated to have been roughly about 35 - 85 curies-assuming one gram of ash had about one curie of radioactivity at the time it fell on the boat.

When the radioactive dust is scattered uniformly on a plane surface which may be considered large enough as compared with the mean free path of the gamma-ray photon, the gamma-ray flux (I_γ) received at a height "h" above the ground may be given approximately by the following expression,

$$I_\gamma = \frac{S}{2} \int_k^\infty \frac{\exp(-kR)}{R} = dK = -\frac{S}{2}\left[E_i(-kh)\right] \qquad (2)$$

where S is the total energy of gamma-ray emitted per unit area per unit time (Mev/cm^2 -sec), k the linear absorption coefficient (1.0×10^{-4} cm^{-1}).

Table 2

Patient	Sample	Activity*	Date collected from the patient
K	blood	$10^{-4}\sim10^{-5}\mu c/cc$	March 17
A	feces	$2\times10^{-2}\sim10^{-3}\mu c/gm$	March 15~17
T	urine	$10^{-3}\sim10^{-4}\mu c/cc$	March 15~17
O	urine	$10^{-5}\sim10^{-6}\mu c/cc$	March 17
S	urine	$10^{-5}\sim10^{-6}\mu c/cc$	March 17
**	hair	$0.1\sim1\mu c/gm$	March 16

* Because of the considerable differences detected in the different aliquots of the sample, only the range of activity as estimated with the substandard source of Sr^{90}-Y^{90} is given. Most probably this is partly due to the fact that some of the radioactive nuclides were lost by the absorption or deposition on the metallic part of the syringes or on the walls of the container while the samples were transfered from one container to another for transportation. Some samples had been diluted for various clinical tests before we received them at the hospital and the degree of dilution was not clear.

** ~~This hair was~~ not specifically identified.
Whose hair it was

When the radioactive contamination is x c/m^2, S may be expressed by

$$S = 3.7 \times 10^6 E_\gamma X \ MeV/cm^2 - sec. \qquad (3)$$

where E_γ is the mean energy of gamma-rays (0.7 Mev).

The mass absorption coefficient of the gamma-ray with the energy 0.08 - 2.5 Mev may be considered approximately constant to about ten percent. For the gamma-ray with this range of energy, the incident beam of gamma-ray which corresponds to 1 rad may be given by

$$I = \frac{2.3 \times 10^9}{E_\gamma} photons/cm^2 \left(\fallingdotseq 2.3 \times 10^9 \ MeV/cm^2\right) \qquad (4)$$

Therefore the dose rate d(rad/hr) at a height of about one meter above the ground may be expressed by the following e equations.

$$\alpha = -\frac{3.7 \times 10^6 E_\gamma \times \left[E_i(-kh)\right]}{2 \times 2.3 \times 10^9} \times 3,600 \ rad/hr \qquad (5)$$

With the above equation the dose rate corresponding to a radioactive contamination of 1 c/m^2 may be estimated about 8.25 rad/hr. Assuming the gamma activity to be roughly about one half of the beta activity the dose rate corresponding to the beta activity of 35 - 85 c/m^2 may be estimated to be roughly about 150 - 350 rad/hr. Since the area of the deck of the small boat is smaller compared with the mean free path of the gamma-ray photon, the actual dose rate the fishermen might have received on the boat at the time of deposition of the radioactive dust would be smaller than the above value.

Although it seems that most of the dust would have been washed away from the deck of the boat some time after it deposited, the external dose received by the fishermen before they washed

the dust away from the boat may be estimated about the order of 100 - 300 rad. Therefore adding to this the dose due to residual activity on the boat the total external dose the fishermen might have received during two weeks voyage may be estimated roughly about 400 - 800 rad. Judging from these figures it seems probable that more than 50% would have died without proper medical care directly or indirectly due to radiation.

In connection with this it should be noted that the cabin, where Mr. Aikichi Kuboyama the chief wireless operater of the boat who died at 6:56 p.m., Sept. 23, 1954 was living, was located at the highest part of the boat. When the author examined the boat, the roof of the upper deck above his cabin seemed to be the only place in the boat where the dust did not seem to have been cleaned thoroughly. Therefore it seems very probable that he would have been one of the crew who recieved the highest external gamma-ray dose during the voyage although the degree of his skin injury due to the direct contact of the radioactive dust appeared to be smaller than some of other fishermen.

In view of these findings, there is good reason to believe that the crew had inhaled or ingested at least some of the radioactive dust during the voyage in addition to receiving external radiation.

3. Alpha-ray Detection

Because of the harmful effects of alpha-rays, it was very important, especially from the medical or health-physics point of view, to determine whether or not any alpha-emitters may have been included in the Bikini ash. Therefore, a measurement of alpha-rays was attempted, using windowless ionization chamber of conventional type, with liner amplifier and pulse height selector which had previously been calibrated by P_o-α.

Towards the end of March 1954, a fraction which was thought to contain the transuranium elements was separated from the strongly radioactive original sample of Bikini ash of about 10^5 cpm beta activity, according to the peroxydisulfatefluoride method(24) From this fraction alpha-ray activity of about 105 counts per 30 minutes was detected above natural, which seemed to be at least partly due to the possible existence of long lived Pu^{239}. However, there seemed to be a decaying component due to some other nuclides also. The dimensions of the ionization chamber used for this alpha-ray detection are shown in Fig.5 in units of cm.

(Fig 5)

From this transuranium fraction, beta-ray activity about one hundred times stronger than alpha activity was detected when the fraction was separated towards the end of March 1954. However, the exact confirmation of nuclides could not be done because of the difficulties in separating transuranium elements from each other quickly.

It was necessary to estimate the beta-ray energy more accurately from the health physics point of view, and the beta-ray analysis was conducted in cooperation with Prof. Azuma and his staff of the Faculty of Engineering, Osaka Prefectural University.

4. Beta-ray Analysis

4.1 Experimental Procedure

The continuous beta-ray spectrum and the internal conversion lines from the radioactive elements were measured using a double-coil magnetic-lens beta-ray spectrometer. First, the beta-ray spectrum of the original sample prior to chemical analysis was studied on March 19. The radioactive ash was put on a mica sheet of about 3 mg/cm^2 and covered with Zapon film of about 30 $\mu g/cm^2$

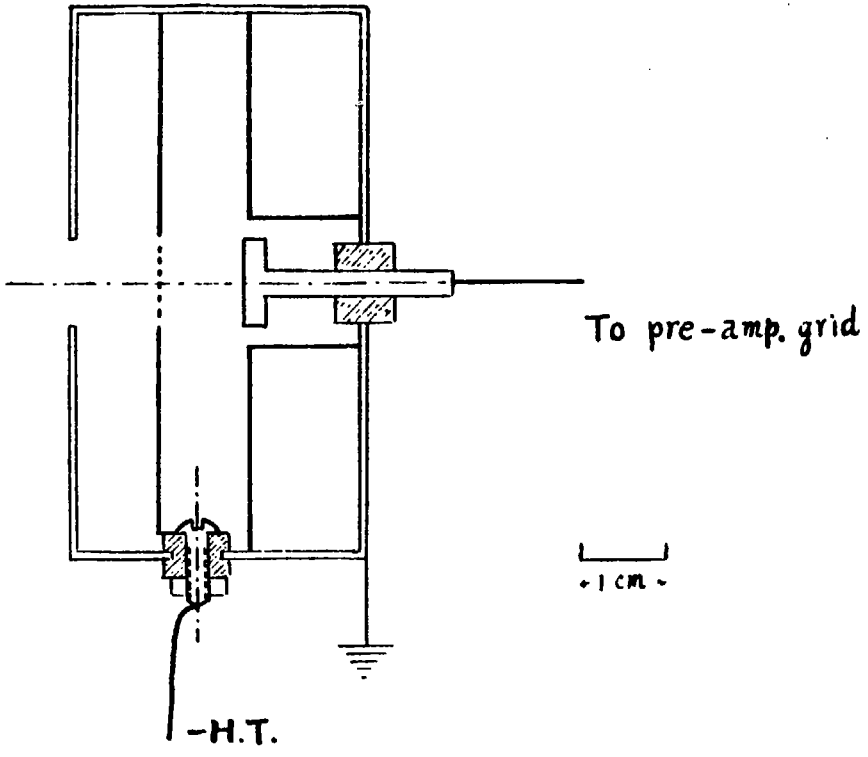

Fig. 5. Ionization chamber for alpha-ray detection.

to prevent its dispersion. The observed beta-ray spectrum is shown in Fig.6. However, there were so many groups of beta-ray and gamma-ray that they could not be analyzed reliably without chemical separation. The ash was then analysed by the usual method of rediochemical analysis[1)(19)(20)] A strong beta-ray activity was found in the CeO_2-fraction (about 40 - 50% of β-activity of rare-earth elements mixture). The beta-ray analysis of this fraction was attempted. The chemically separated CeO_2-fraction in powder form was put on a mica sheet and covered with a thin Zapon film from the same source as for the beta-ray analysis.

(Fig.6)

The beta-ray spectrometer used in the work was the ordinary double-coil magnetic-lens type. The spectro-chamber was equipped with an aluminium baffle system like those of Van Atta et al[(7)] to reduce the scattered electrons. The beneficial effect of this baffle system on the measured spectrum was investigated with the internal conversion line of the 665-kev gamma-ray from Cs^{137}[(8)]. The baffle system of the spectro-chamber is shown in Fig.7 in units of cm.

(Fig.7)

In operating the spectrometer, a resolution of about 5 percent was used to increase the transmission. The detector consisted of an ordinary G-M counter of the end-window type, whose mica sheet was 2.9 mg/cm^2 thickness.

The measured beta-ray spectrum was analyzed using the Fermi plot; that is, the plot of $(N/f(z,w))^{1/2}$ against the total energy, where N is the number of counts per minute of beta-ray per unit momentum, and $f(z,w)$ is the Fermi distribution function of transition probability, and w is the energy of emitted electrons in units of mc^2. In this analysis a better approximation for $f(z,w)$ especially for large z, as given by Bethe and Bacher[(9)] was used.

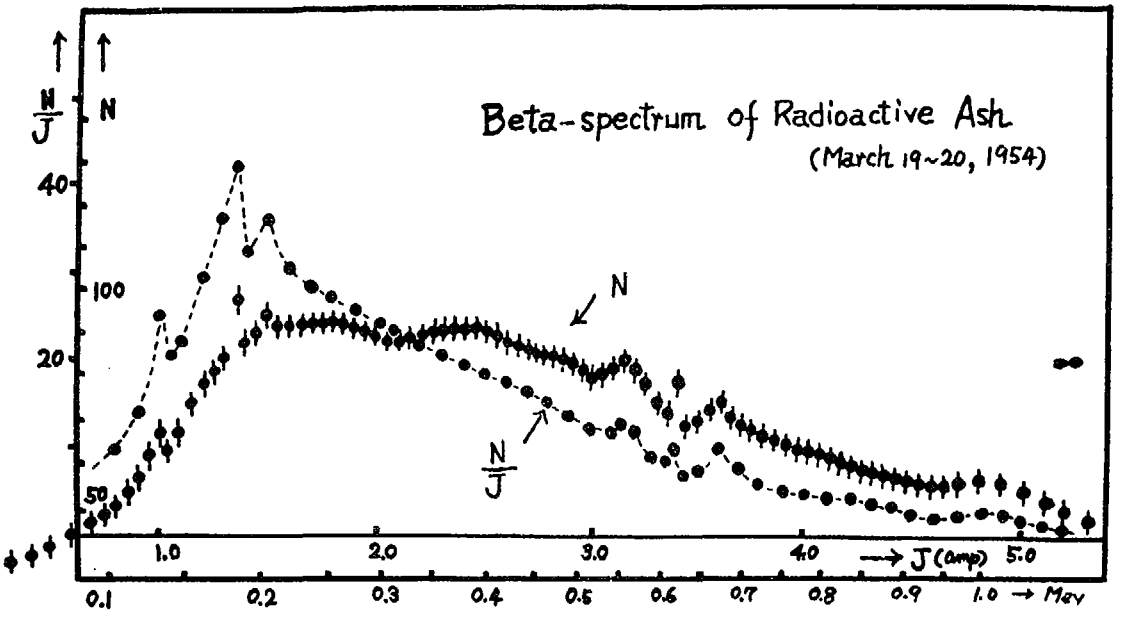

Fig. 6. Beta-ray spectrum of the original radioactive ash prior to chemical analysis.

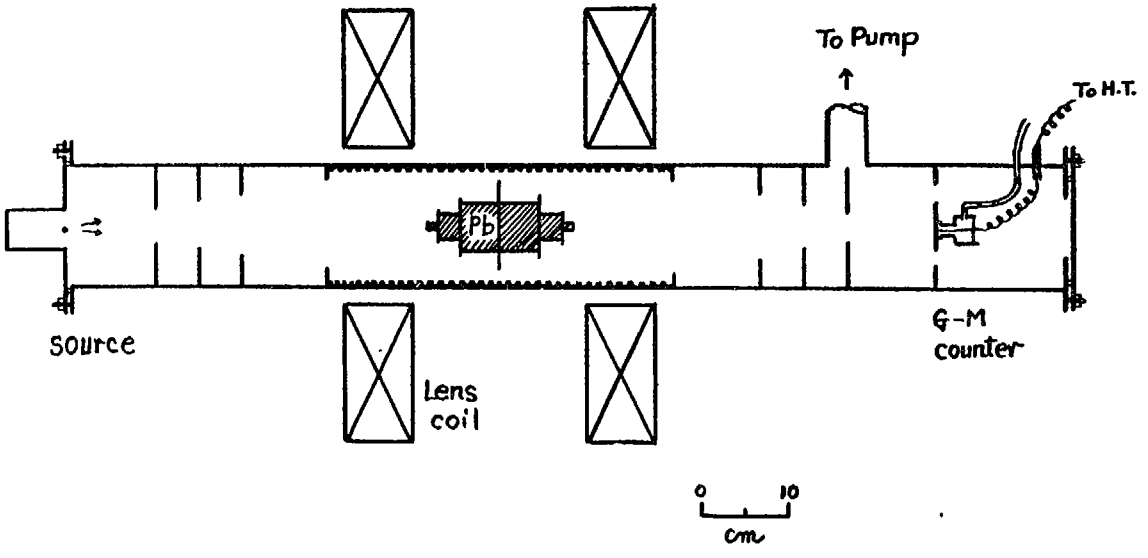

Fig. 7. The baffle system of the spectro-chamber.

4.2 Results and Discussion

The absorption curve of the CeO_2-fraction prior to beta-ray analysis is shown in Fig.8. In Fig.9 the absorption curve of this fraction on April 19 is compared with that on May 15, from which the apparent half-life of the higher energy part of beta-ray and that of the lower energy may be estimated separately.

In Fig.10 is shown the observed beta-ray spectrum of the CeO_2-fraction. The insert in Fig.10 shows the internal conversion line of the 134-kev gamma-ray. The allowed Fermi plot of the higher energy part of the beta-ray is shown in Fig.11, from which the end-point energy of 2.97-Mev can obtained. This beta-ray group was then subtracted, and the remaining beta-ray groups were also analysed using the Fermi plot. It can be seen that the measured beta-ray group is a complex consisting of six components with end points at 1.377-, 1.086-, 1.030-, 0.709-, 0.580-, and 0.438-Mev.

(Fig.8)
(Fig.9)
(Fig.10)
(Fig.11)

The data obtained in the present work were compared with the momentum spectra of pure Ce-isotopes which have been reported hitherto. Porter, Cook, and others[10]-[12] have measured the beta-ray groups of pure Ce^{144} (289 d) -Pr^{144} isotope with a lens-coil spectrometer, and have observed the beta-ray groups of 2.97-, 0.30-, and 0.17-Mev, and the gamma-ray groups of 134-, 100-, 80.7-, 54-, and 33.7-kev. For Ce^{141}, Feedman, Engelkemeir, and others[13]-[15] have reported the beta-ray groups of 581-, and 442-kev, and the gamma-ray group of 143-kev. For Ce^{143}, Burgus and others[16]-[18] have measured the beta-ray groups of 1.39-, and 1.09-, and 0.71-Mev, and the gamma-ray groups of 720-, 660-, 356-, 289-, 160-, and 35-kev.

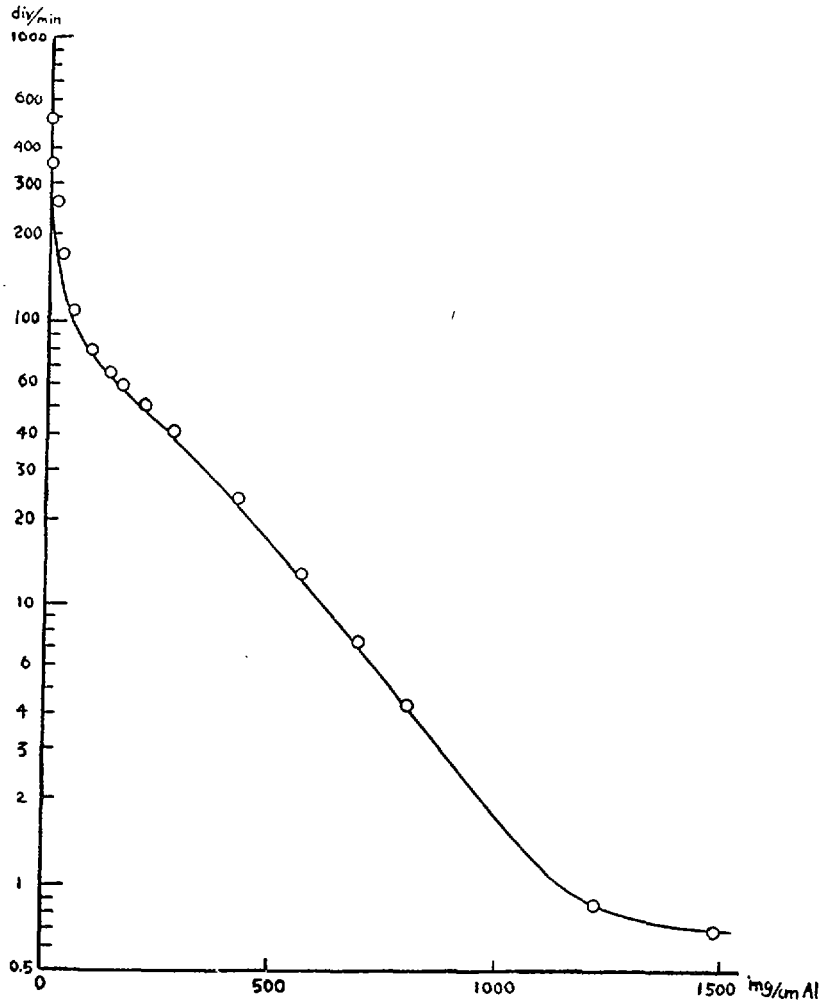

Fig. 8. The absorption curve of the CeO_2-fraction isolated from Bikini ash prior to beta-ray analysis. (Lauritsentype electrometer with 3.5 mg/cm² Al window)

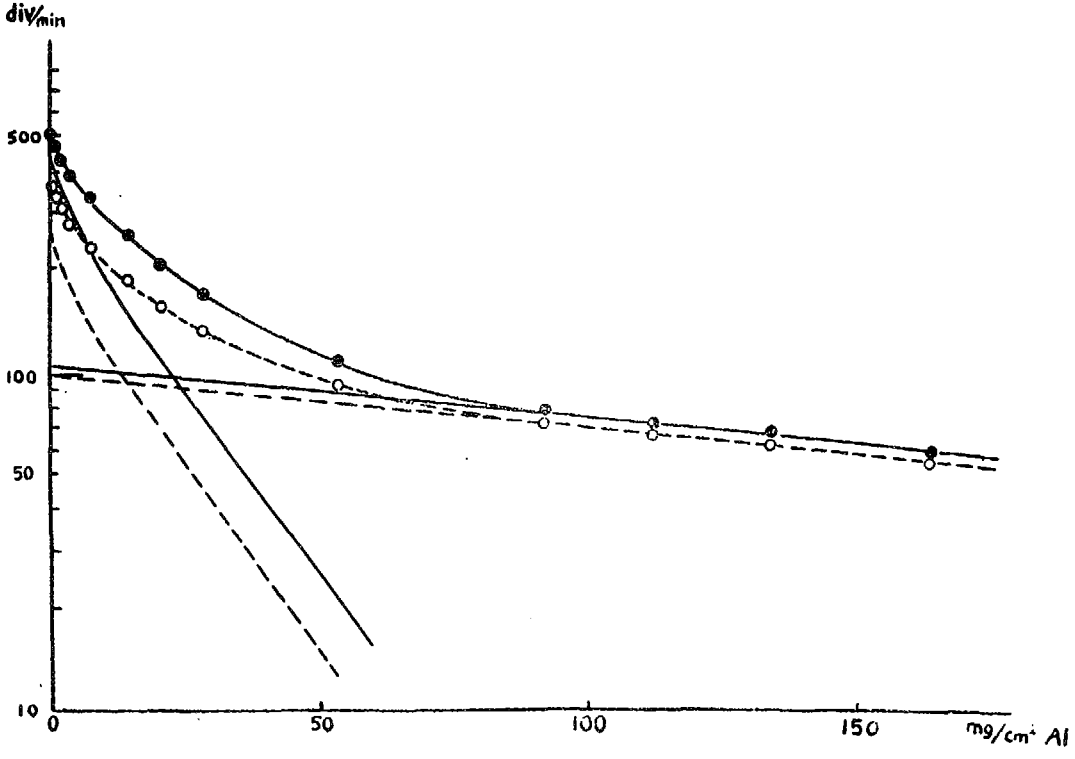

Fig. 9. The absorption curve of the CeO_2-fraction on April 19 (solid line), compared with that on May 15 (dotted line). (Lauritsen-type electrometer with 3.5 mg/cm² Al window)

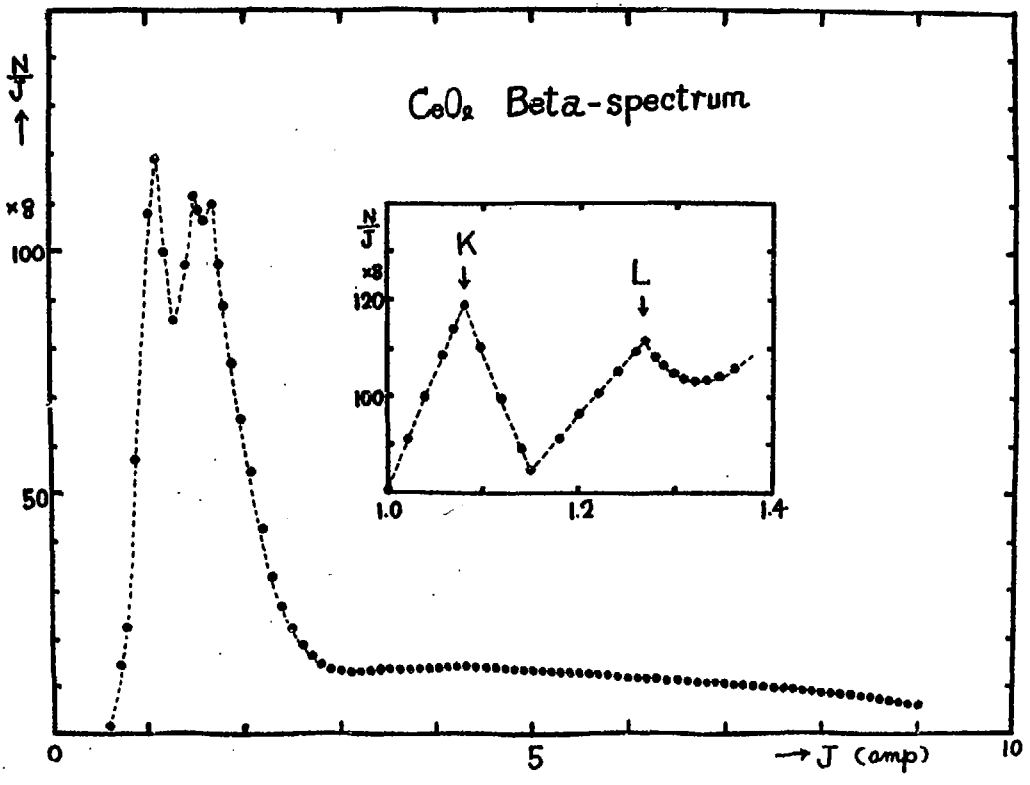

Fig. 10. Beta-ray spectrum of the CeO_2-fraction (The insert shows an enlargement of the internal conversion line of the 134-kev gamma-ray from Ce^{144}-Pr^{144}.)

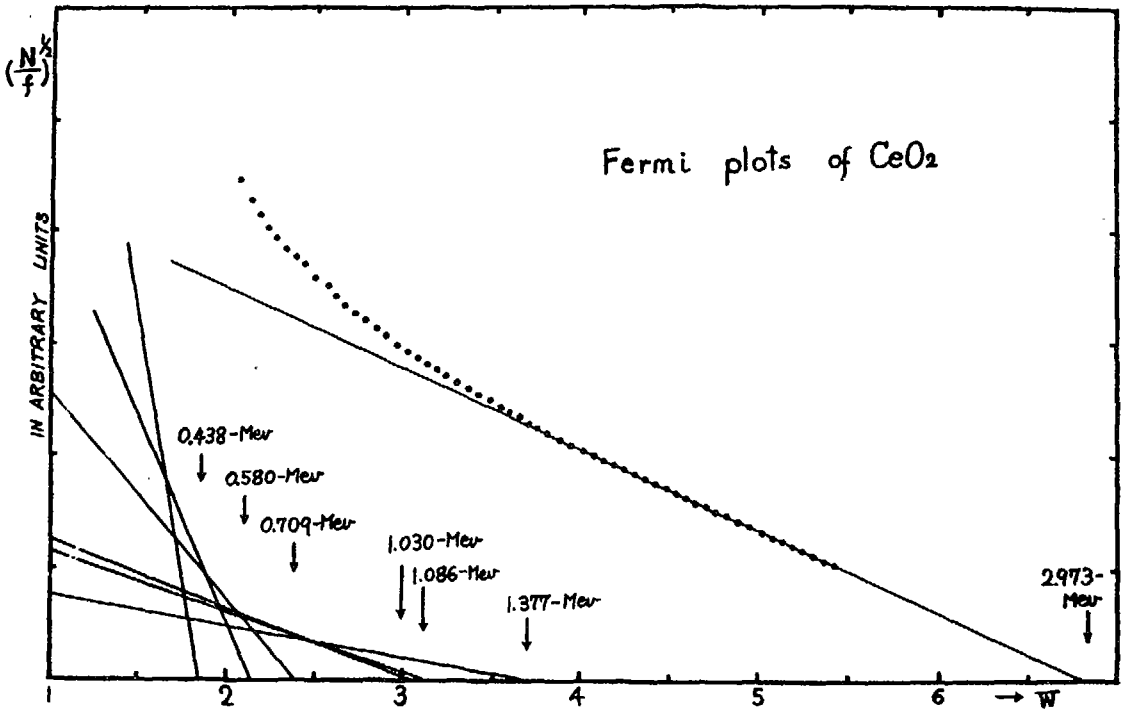

Fig. 11. Allowed Fermi plots constructed from the beta-ray spectrum of the CeO_2-fraction (The values of the maximum energies of the beta-ray groups are shown in the figure.)

In this experiment, Ce^{144}-Pr^{144} was ascertained to exist in the sample, from the beta-ray spectrum of the highest energy and the internal conversion line of the 134-kev gamma-ray. The half-life of Ce^{141} is short compared to that of Ce^{144}, but the existence of the former element in lower concentration may be inferred from the beta-ray groups of 580- and 438-kev. The energies of the internal conversion lines of Ce^{144} and Ce^{143} differ from each other by only 10-kev, and they have spectra of similar shape. Therefore, these two could not be detected separately because of the low resolving power of the beta-ray spectrometer used in the present work, and because of a small existing amount of Ce^{143}; but the possible existence of Ce^{143} may be inferred by the beta-ray group of 1.377-, 1.086-, and 0.709-Mev.

Since it is rather difficult to separate the elements Ce and Y completely by ordinary methods of chemical separation(1), a small amount of Y may be contained in the sample on a contamination level. However, the identification of the 1.537-Mev beta-ray group of Y^{91} was difficult in the CeO_2-fraction.

In conclusion, the beta-ray groups corresponding to Ce^{144}-Pr^{144}, Ce^{143}, and Ce^{141} were identified from Fermi analysis of the beta-rays emitted from the CeO_2-fraction which was chemically separated from the ash collected on board the No.5 Fukuryu Maru. The other elements were also chemically separated, but Fermi analysis was not undertaken because of the weaker radioactivities after separation into various fractions. The radiochemical analysis was conducted in cooperation with Prof. Yamatera and his staff of the Institute of Polytechnics, Osaka City University.(19)(20)

5. Radiochemical Analysis

Since it was urgently required to identify possible radioactive nuclides from the health physics point of view, this radiochemical analysis was conducted with special reference to fission products.

5.1 Outline of the Method of Analysis

The sample was dissolved in dilute nitric acid, and the isotopic or proper nonisotopic carriers were added when the solution was analysed according to the ordinary method of systematic qualitative analysis. The following findings have been obtained from the preliminary experiment conducted during the initial 24-hour period after obtaining the sample.

(1) Part of the radioactivity of the "ash" was found in the portion which was precipitated by hydrogen sulfide from the acid solution.

(2) A greater part of the radioactivity of the "ash" was found to collect in the portion which was precipitated by ammonia and ammonium sulfide in the presence of ammonium chloride. Nearly all of this part of the radioactivity was also precipitated by ammonia alone. More than one-half of the activity was precipitated by oxalic acid from weak acid solution.

(3) Part of the radioactivity was also found in the portion of alkaline earth metals.

Such a distribution of radioactivity as in the above was similar to that of fission products. Therefore the analytical procedure was modified to some extent for convenience's sake to detect the possible radioactive nuclides. Thus the iodine was precipitated as silver iodide together with cations of group I. And the

portion which may be precipitated by hydrogen sulfide from acid solution was separated into two groups; the one which may be precipitated from strong acid (ca. 2N) solution (carrier: Sb_2S_3) and the other which may be precipitated from dilute acid (ca. 0.3N) solution (carrier CdS). With this procedure a considerably good separation of ruthenium and tellurium seems to have resulted unexpectedly. The hydroxides which were precipitated by ammonia in the presence of ammonium chloride were treated with special attention to the rare earth elements and zirconium.

When two different radioactive nuclides co-exist and one of them has a shorter half-life, the half-lives were estimated from analysis of the decay curve. (half-life of I^{131} and I^{132} in $I^{131}+I^{132}$).

When the beta-ray energies of two coexisting nuclides differ greatly, the higher energy part may be estimated separately with a proper absorber. The intensity of the lower energy beta-ray may also be estimated as the difference between the intensity of the higher energy beta-ray and that of the total beta-ray. By this method the half-life of each of the nuclides may be estimated separately for some nuclides without waiting for the one with shorter half-life to decay completely. (half-life of Ru^{105} and Ru^{106} in $Ru^{103}+Ru^{106}+Rh^{106}$, half-life of Ce^{144} in $Ce^{141}+Ce^{144}+Pr^{144}$).

Besides the beta-ray analysis with beta-ray spectrometer as reported in previous section, the maximum energy of beta-rays of individual nuclides was approximately estimated from the absorption curves.

5.2 Analysis of Fission Products

(a) Sr^{89}

Sr^{89} was separated from barium and calcium by the K_2CrO_4 method.[5] The shorter half-life observed may be due to some

contamination by such nuclides as Ba^{140}. Therefore, the estimated percentage of Sr^{89}, 1.6% as of March 26, may include some error although correction was made for the probable amount of contamination.

(b) $Sr^{90}+Y^{90}$

A solution containing lanthanum as a carrier of Y^{90} was added to a solution containing strontium free from other metals, which was obtained from another portion of a large quantity of the original sample. From this solution, hydroxides of rare-earth metals were precipitated. These were then dissolved in dilute acid and converted into oxalate by precipitating with oxalic acid.

The oxalate containing Y^{90} had an activity which may be estimated to be about one hundredth of the total activity of the strontium portion. This value for Y^{90} activity may be considered to be an underestimate, since the oxalate was separated from the solution before its complete precipitation.

(c) $Zr^{95}+Nb^{95m}+Nb^{95}$

Hydroxide precipitate by ammonia resulting from the ordinary analytical procedure was dissolved in HCl (1 : 10) and the phosphate of zirconium was precipitated with Na_2HPO_4. The shape of the decay curve which was flat at the end of March and began to show a decreasing tendency from April to May with the apparent half-life of about 70 days, may be explained by assuming that a large part of niobium might have been precipitated with the zirconium.

(d) $Ru^{103}+Ru^{106}+Rh^{106}$

Nuclides of ruthenium are considered to have been carried with antimony sulfide upon its precipitation from a strong acid solution, about 2N in HCl and HNO_3. Thus, toward the end of

April, β-rays from this antimony sulfide precipitate had a rather simple absorption curve which may be interpreted by assuming that the main components of β-rays consist of the one with high energy from Rh^{106}, daughter of Ru^{106}, and another with low energy from Ru^{103}. Moreover the half-life of each nuclide measured seemed to support this interpretation.

(e) Te^{129m}+Te^{129}

It was shown by the β-ray absorption curve which was measure on April 24, of the sulfide portion coprecipitated with cadmium sulfide carrier from 0.3N acid solution that the main component of the β-rays was the one with a maximum energy of 1.8 Mev. The apparent half-life of radioactivity of this portion, including some nuclides with shorter half-lives was about 20 days. Though identification of the nuclide with 1.8 Mev β-ray was not successful on account of inadequate chemical treatment, this nuclide is likely to have been Te^{129} accompanied by its parent Te^{129m}.

(f) Te^{132}+I^{132}+I^{131}

Radioactive iodine was separated from cations other than group I by coprecipitation with silver chloride. The presence of I^{132} and I^{131} was confirmed from the analysis of the decay curve and the absorption curves at different times. Te^{132} was deduced to be present from genetic relationship.

(g) Ba^{140}+La^{140}

Barium was separated from strontium by the K_2CrO_4 method.[22] A single treatment according to this method could not result in complete separation. However, the presence of Ba^{140} could be confirmed, since weaker activity of Sr^{90}, parent of Y^{90}, made it easy to confirm the formation of La^{140} from Ba^{140} through chemical separation of La^{140} from its parent.

(h) Ce^{141}+Ce^{144}+Pr^{144}

Mixed compounds of rare earth elements, which were obtained from another large quantity of "ash", were treated according to the iodate method[23] A doubly repeated treatment resulted in a good separation of cerium from the other rare earth elements. The presence of Pr^{144}, daughter of Ce^{144} was confirmed on the basis of β-ray absorption curves of cerium portion. Since the apparent half-life of the activity of low energy β-ray of this portion, β-ray of Pr^{144} is of high energy, was found to be 36 days from the analysis of the absorption curves of beta-rays at different times between April 19 and May 20. From comparison of this observed half-life and the known half-lives of Ce^{141} and Ce^{144}, the ratio 4:1 was deduced for the ratio Ce^{141}:Ce^{144} disintegration rate on April 19. The value 7:1 is calculated as the ratio on March 26.

The beta-ray analysis of this fraction with the beta-ray spectrometer was also made and the activity of Ce^{144} was estimated to be approximately 2% of the total activity of the mixture of rare earth elements as of March 26.

5.3 Other Findings and Discussions

Since it was known that the "ash" consisted mainly of calcium hydroxide, calcium was examined for radioactivity. In calcium, activity less than 2% of Sr^{89} activity was found, but the greater part of it was considered to be due to contamination with Sr^{89}. In this respect, we are not consistent with K. Kimura et al[2] who found Ca^{45} in such a high activity as about 20% of Sr^{89} activity. This inconsistency may be due to the large self-absorption of low energy β-ray of Ca^{45} with a lar e amount of the carrier.

Later, in November 1954, a radioactive nuclide which may be considered to be Zn^{65} was detected from the intestine (ca. 47% activity) and the ovarium (ca. 18% activity) of the tuna

fish caught in the latter half of 1954 in the Pacific.

The absorption curve of the radiation emmitted from this nuclide was observed to agree well with that of Zn^{65} obtained from Oak Ridge. However, this nuclide was not found in appreciable amount in the "Bikini Ash" collected from the Fukuryu Maru.

From an aliquot of the original sample of Bikini Ash reserved for about two years for the purpose of detection of long life fission products, strontium was separated from calcium with the concentrated nitric acid method after the addition of carrier Sr. Waiting for about 20 days for the growth of Y^{90} after the separation and purification of Sr, Y-fraction was separated from Sr with the ion exchange method (Dowex 50) after the addition of carrier Y. The existence of $Sr^{90}+Y^{90}$ was confirmed by the detection of Y^{90} beta-activity in this Y-fraction.(26) $Cs^{137}+Ba^{137m}$ was also confirmed by the detection of photo-peak corresponding to this nuclide with the gamma-ray scintillation spectrometer in the cesium bismuth iodide precipitate(26) from the alkaline metal fraction with carrier Cs. The ratio of activity of Sr^{90} to that of Cs^{137} was estimated to be about 1:1.7. The radioactive free samples of Sr, Y, and Cs used as the carrier in this analysis were kindly provided to the author from Prof. Nishi of Kyoto University.

Judging from the various conditions of the local radioactive fallout on the boat 80 - 90 miles away from the Bikini Atoll and assuming the surface radioactive contamination of 50 curies/m^2 to be a representative average figure for the elliptical area of about 200 km downwind distance with the maximum cross wind width of 60 km as deduced from the comparison with the mode of spreading of volcano ash in the past, the total activity in this area may be estimated to be roughly about the order of magnitude of 7×10^{11} curies with higher degree of contamination closer to the test

site and lower degree away from the site. Assuming that roughly about one half of the total fission products fell in the local area and that the activity per one gram of fission products at a few hours after fission is about three million curies, the total amount of fissile material that underwent fission at the time of nuclear detonation may be estimated at about 4.6×10^5 g or roughly about 500 kg.

Comparing the above estimation with the atomic bombs dropped over Hiroshima and Nagasaki which are assumed to correspond to 20 Kilotons TNT equivalent detonations with the nuclear fission of about 1 kg of U^{235} in case of Hiroshima and of Pu^{239} in case of Nagasaki, the size of the thermonuclear test conducted in Bikini on March 1, 1954 may be estimated to be about 10 megatons TNT equivalent detonation.

However, since such a large amount of nuclear fission as estimated from the high degree of radioactive contaminstion of No.5 Fukuryu Maru seemed to be almost impossible with the slow neutron fission of U^{235} or Pu^{239} because of the limitation due to critical mass, the author was led to a conclusion[34] that the nuclear detonation in Bikini must be due to the fast neutron fission of a large amount of natural uranium or U^{238}.

Later this assumption was supported by the detection of uranium 237,[2][5][29] which was reported by K. Kimura, et at[2] to have existed in about 20% of the total activity as of March 26, 1954. Moreover, the ratio 1:1.7 of the activity of Sr^{90} to that of Cs^{137} seems to be closer to that of the fast neutron fission of uranium 238(1:1.9 - 2.0)than to that of the slow neutron fission of uranium 235(1:1 - 1.2).

In view of these findings in the light of the present knowledge of nuclear physics and engineering it may be inferred that the fast neutrons which caused the fast neutron fission of a large quantity of natural uranium or U^{238} would have been derived from some fusion reaction initially ignited by a slow neutron fission of U^{235} or Pu^{239}, as pointed out by R. E. Lapp,[31][34] J. Rotblat,[32] and Indian scientists.[33]

6. Estimation of Internal Dose

In order to obtain a clue to estimating the possible internal dose of radiation due to ingestion or inhalation of the Bikini Ash, we have conducted, from the health physics point of view, a series of biological experiments by administering the extract of the dust collected from No.5 Fukuryu Maru to about 100 rats during the period from late March to May, 1954.

As a result, radioactive iodine was observed to accumulate rapidly in the thyroid gland. Therefore, we obtained the impression that the selective power of uptake of specific elements by some of the organs of the animals might help as a subsidiary measure of the rapid radiochemical identification of some of the nuclides in case of emergency.

After autopsy, the radioactive decay of different organs have been measured. However, since the extract of the dust administered to the animal was a mixture of various radioactive nuclides, the radioactive decay did not seem to obey a simple exponential type, therefore we have tried to approximate it by a power function of the time t after the nuclear detonation on March 1 with the negative exponent of t. The general expression may be given by

$$A = A_1 t^{-n} \qquad (\epsilon)$$

where A is the activity in the organ excised from the experimental animal at time t after the nuclear fission, and A_1 and n are constants.

With most of the organs except the thyroid gland, the average value of n or the exponent of t was observed to be roughly about unity ranging from 0.8 to 1.3, with somewhat lower value with the bone occasionally. However with the thyroid gland the value of n was observed to be roughly about 2, or sometimes it was estimated to be as high as 2.37. A part of the reason might be ascribed to

the higher percentage of activity due to radioactive iodine I^{131} in the thyroid gland during this period of observation. Some of the examples are shwon in Figs.12 - 13. Among the various complex

(Fig.12)

(Fig.13)

factors involved in this type of analysis, we must pay attention to the fact that various radioactive nuclides included in the fission products undergo radioactive decay and change into different nuclides with time as shown in Fig.14. Therefore, sometimes

(Fig.14)

there seems to be a possibility that the organ, in which major part of activity may accumulate, might be expected to change with time, and that depending on the time, at which the fission products mixture was administered, somewhat different results may be obtained in the retention ratio as well as in the relative distribution of the radioactivity in various organs.

The estimation of the percentage of retention or uptake of the activity by various organs was attempted, but it was observed to be considerably different with the possible impurity included in the dust, from which the radioactivity was extracted, and the acidity of the extract at the time of the administaration to the animal, and in some extreme cases it was observed to be different by about one order of magnitude. However, as a general tendency, a higher percentage of retention was observed with the extract of lower pH. Some of the results of the experiments, with which relatively higher percentage of retention was obtained, are shown in Figs.15 - 17.

(Fig.15)

(Fig.16)

(Fig.17)

From the analysis of these data, the effective biological half life was estimated to be roughly about 10 - 15 days for the liver and 30 - 100 days for the bone.

THYROID

$A = A_1 t^{-2.37}$

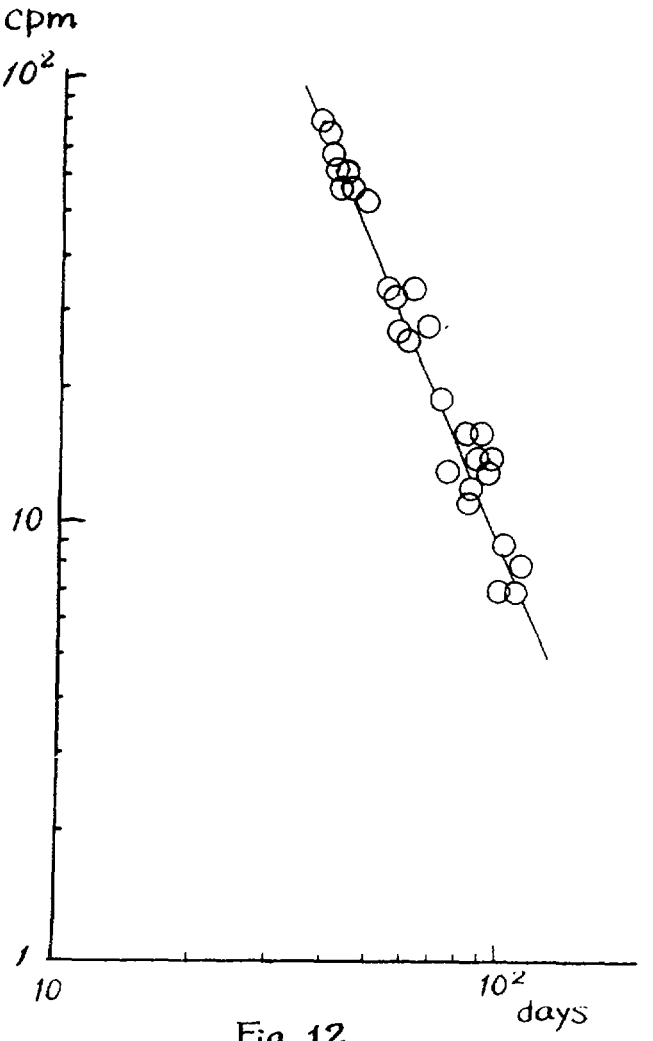

Fig. 12

MUSCLES

$$A = A_1 t^{-1}$$

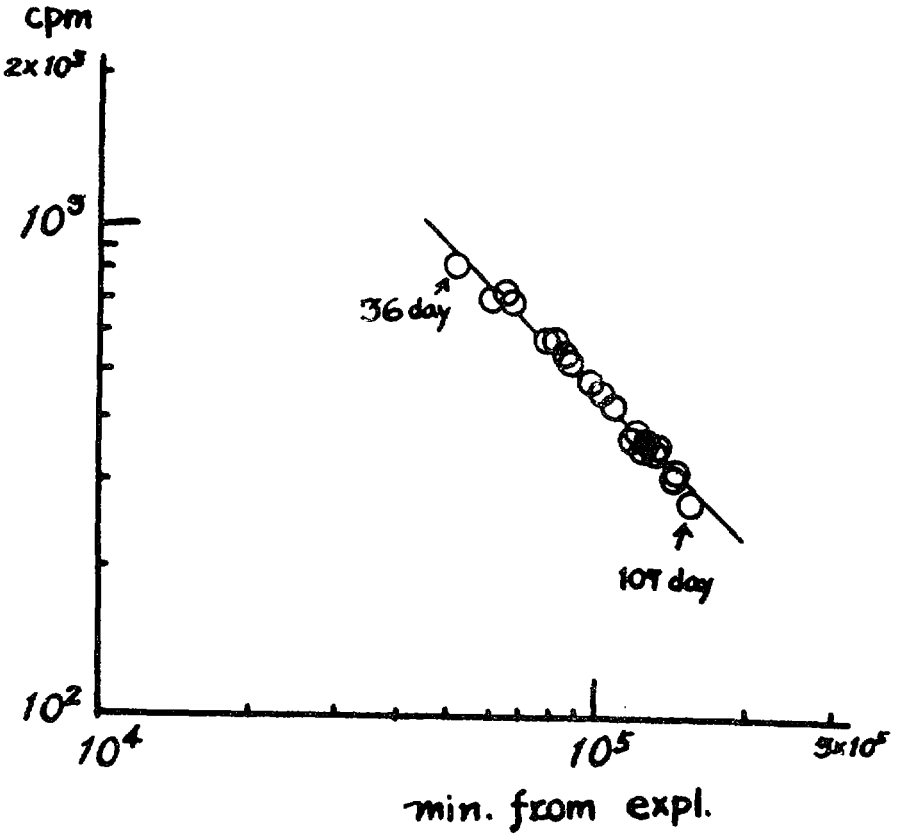

Fig. 13

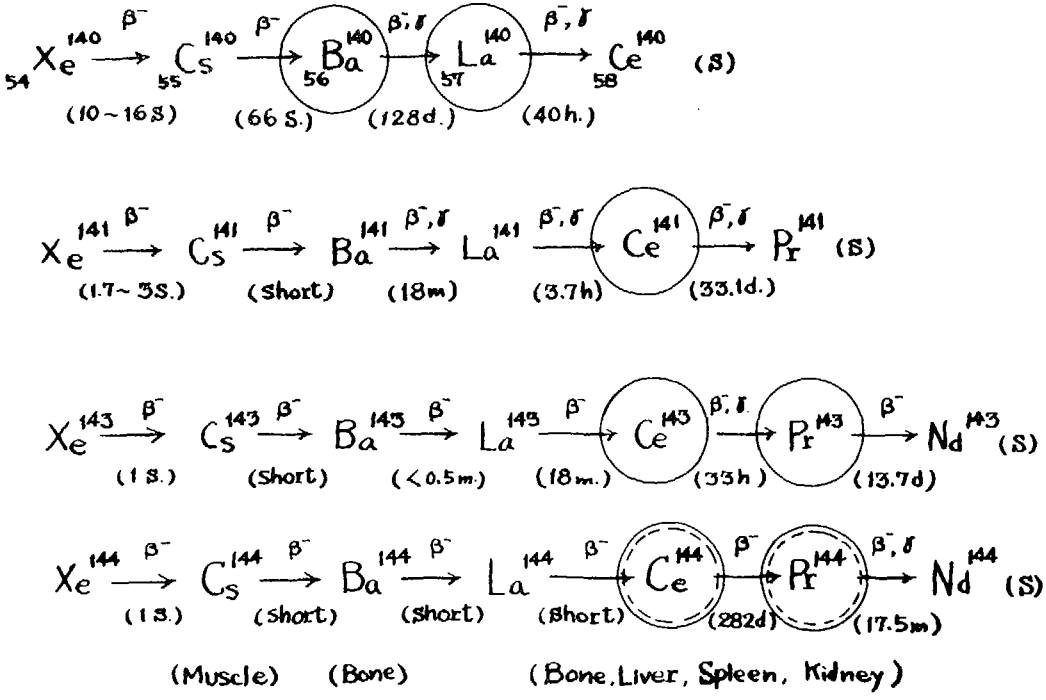

Fig.14 : The nuclides encircled by the solid line are the ones detected in Bikini ash. Those encircled by the broken line are the ones detected in the organs of Mr. Kuboyama at 206 days after nuclear detonation.

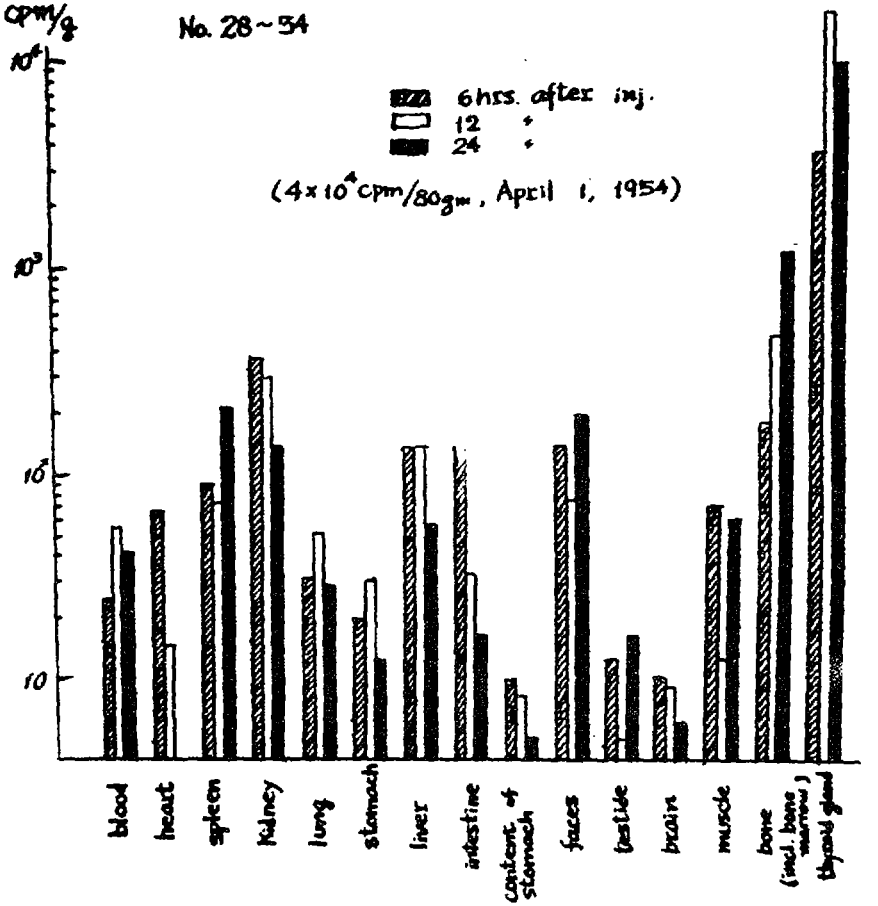
cpm/g
No. 28~34
10^4
10^3
10^2
10
6 hrs. after inj.
12 "
24 "
(4×10^4 cpm/80gm, April 1, 1954)
blood
heart
spleen
Kidney
lung
stomach
liver
intestine
content of stomach
faces
testicle
brain
muscle
bone (incl. bone marrow)
thyroid gland

Fig. 15

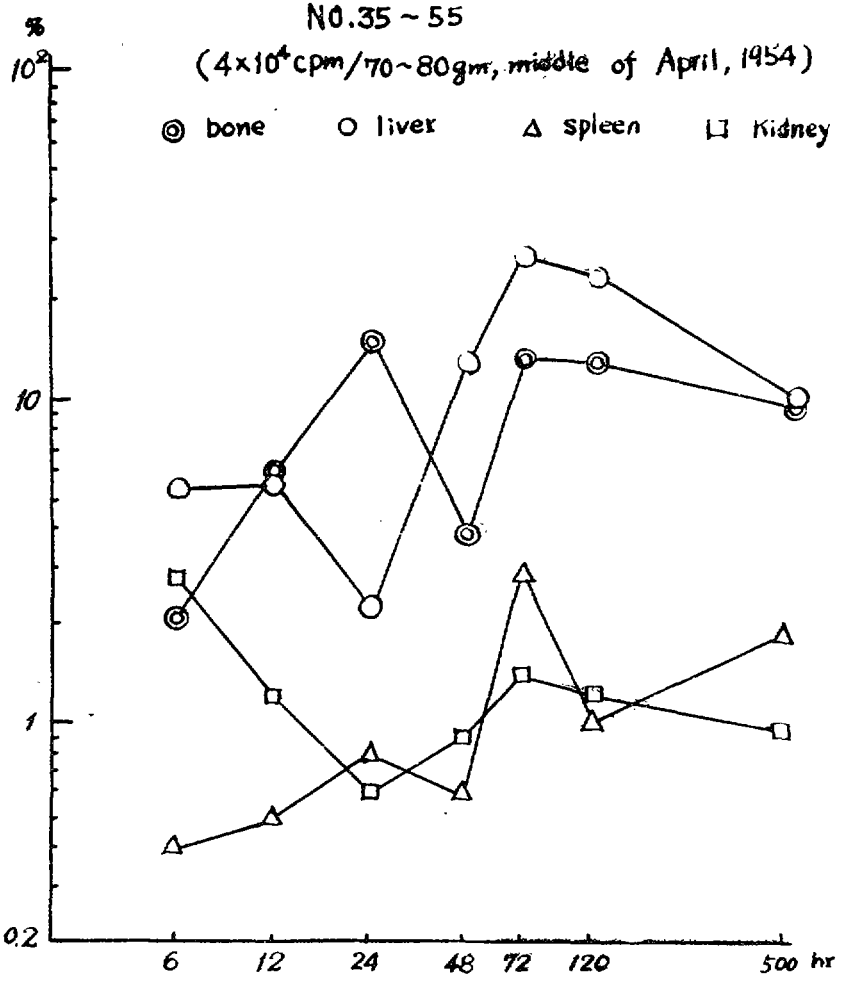
No.35 ~ 55
(4×10⁴cpm/70~80gm, middle of April, 1954)
bone
liver
spleen
Kidney
%
10²
10
1
0.2
6
12
24
48
72
120
500 hr

Fig. 16

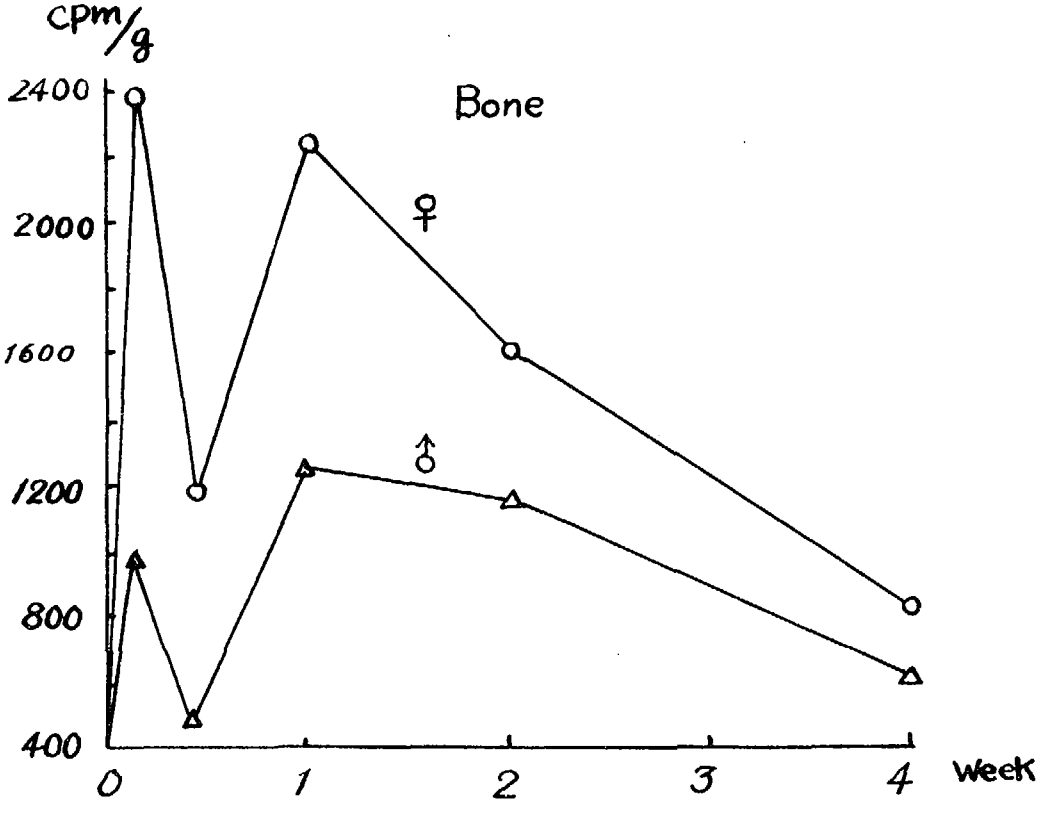

Fig. 17

In view of these findings, it seemed almost impossible to estimate accurately the possible internal dose of radiation the crew might have received during their voyage or at the time the dust fell on the boat. There seemed to be too many unknown factors which might influence the results, such as the acidity or pH in the stomach, whether some of the radioactive nuclides in the dust reacting with the acids in the stomach formed some soluble chemical compounds, or whether some of the nuclides formed some soluble complex with organic acids, or whether some soluble chelate complex of the radioactive nuclides were formed or not, etc. All of these factors seemed to affect more or less the results of the dose estimation, but the most important of all was the difficulty in estimating the exact amount of radioactivity the crew possibly ingested or inhaled on the boat. Although the exact amount was not known, according to the statements of the fishermen, some of the crew seemed to have ingested considerable radioactivity at the time of tasting the strongly radioactive dust to see what it was without knowing that it was strongly radioactive.

Judging from these statements of fishermen and from the degree of radioactive contamination of food and drinking water the author detected in the boat, a rough order of magnitude guess was made as to the amount of radioactivity some of the most highly contaminated crew might have ingested and inhaled on the boat and the upper limit was estimated to be in the range from a few curies to about a tenth of a curie.

In spite of these difficulties, when Mr. Kuboyama, chief wireless operator of No.5 Fukuryu Maru died at 6:56 p.m. Sept. 23, 1954, the author was requested by Dr. Miyoshi of Tokyo University Hospital, who was then the chief doctor in charge of the Bikini patients in Tokyo, and Dr. Kumatori, who was then the

chief doctor in charge of Mr. Kuboyama at the National First Hospital in Tokyo, to estimate the possible internal dose of radiation.

The pathological findings according to the autopsy of Mr. Kuboyama conducted by Dr. Ohashi, the chief pathologist at the hospital may be introduced briefly in the following.

The scars of radiation burns were observed on the neck, left leg and both insteps. The weight of the liver was 860 g, about two thirds of the normal weight of 1,200 g. On macroscopic examination it appeared to be in a stage towards subacute liver cirrhosis. Because of the marked sign of jaundice due to disturbances of liver function, it has been suspected that this might be due to the serum hepatitis through blood transfusions, but it seems to be extremely difficult to prove this definitively in this case. Even if we admit that it was the serum hepatitis, the radiation did not seem to be excluded from the possible causes leading him to death, because of the possibility of the latent virus being activated by radiation, while the resistance of the body against various infections being lowered.(29)

Besides the liver, various pathological changes have been observed in the lungs, bone marrow, kidney, spleen, pancreas, testis, etc. Some mycotic infection has been detected in the lungs and gastrointestinal tract. This may be due to the repeated application of various antibiotics. However without the application of antibiotics or blood transfusions it seems to be very probable that the patient might have died much earlier either by sepsis or by hematopoietic injuries as was the case at the time of Hiroshima and Nagasaki.

After autopsy, some of the organs have been sent to Kimura's Laboratory of Tokyo University and to Nishiwaki's Laboratory of Osaka City University for the analysis of radioactivity. The

grossactivity which may be ascribed to the fission products mixture (after elimination of potassium activity) was estimated at Nishiwaki's Laboratory to be in the range of 10^{-6} - 10^{-4} μc/gm at the time of his death, although the activity at different part of the organ was considerably different, sometimes by a factor of 2 - 5 with the highest difference in the lung.

From the radiochemical analysis conducted by K. Kimura, et al, such nuclides as Ru^{106}+Rh^{106}, Te^{129m}+Te^{129}, Zx^{95}+Nb^{95}, Ce^{144}+Pr^{144}, Sr^{89}, Sr^{90}+Y^{90}, were reported to have been detected in the liver, kidney, lungs, muscle, and bone. Although the detected nuclides were limited and the existing amount was small, extrapolating back to the time the dust entered the body the original amount may be estimated to have been much larger. Taking into account the radioactive decay and the possible rate of biological elimination, it did not seem to be probable at all that the nuclides detected after his death were the only ones that had entered the body.(29)

Although various uncertainties involved in this type of internal dosimetry seemed to be too large to permit any accurate estimation, assuming the activity at the time of death of Mr. Kuboyama was 25×10^{-12} c/g for the bone and 5×10^{-15} c/g for the liver, an order of magnitude estimation of the initial amount of activity in the liver and the bone was attempted by the author, taking into consideration of the results of the biological experiments, based upon the following equation.

$$A = A_1 \frac{e^{-\frac{0.693}{T}(t-\theta)}}{t} \qquad (7)$$

where A is the activity per unit mass of the organ (c/g) at time t after the nuclear detonation, θ the assumed time at which the maximum activity reached the organ, T the average biological half life, and A_1 a constant.

(Fig.18)

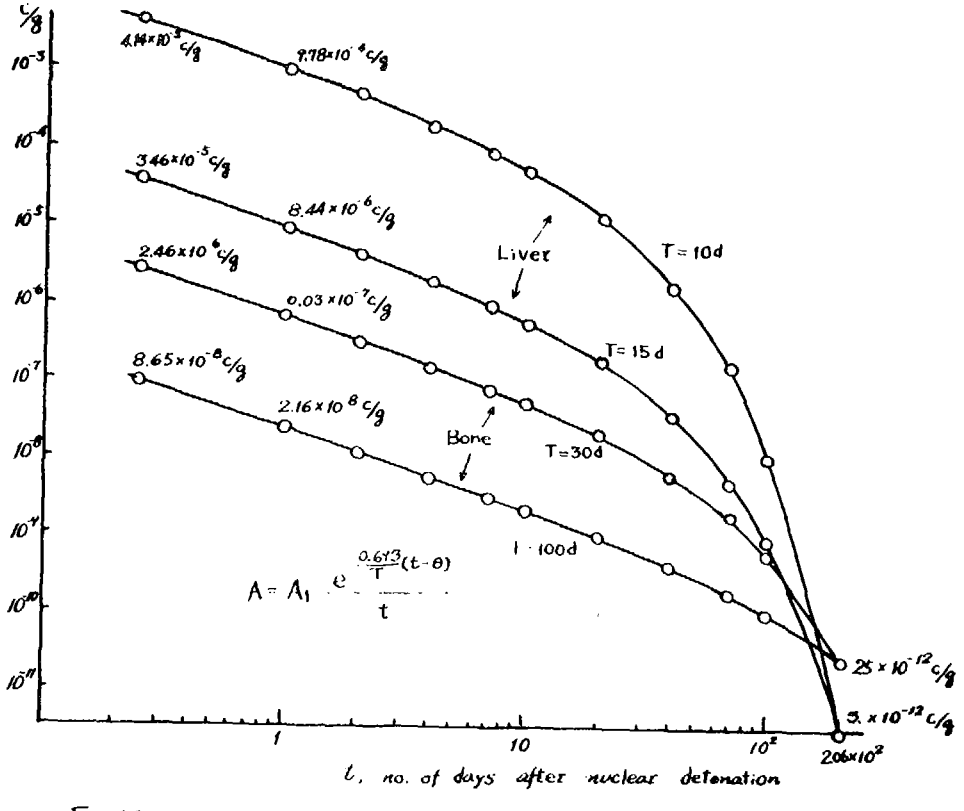

Fig. 18: The activity of liver and bone as estimated from the activity detected in the organs of Mr Kuboyama at 206 days after nuclear detonation.

The results of the estimation of the activities in the bone and in the liver are shown in Fig.18, where the average biological half life T in equation (7) is assumed to be 30-100 days for the bone and 10-15 days for the liver. The corresponding internal dose of radiation in unit of rad for the period from the time θ to the time t_1 after nuclear detonation may be given by the following equation.

$$D_{(\theta,t_1)} = \frac{3.7\times10^{10}\times86400\times1.6\times10^{-6}}{100}\int_{\theta}^{t_1} A_{(t)}\,\bar{E}\,\alpha t \qquad (8)$$

where $A_{(t)}$ is the activity per unit mass of the organ as expressed in unit of c/g at the time t after the nuclear detonation and $\bar{E}$ the effective energy of radiation, and θ and t_1 are expressed in number of days after the nuclear detonation.

Assuming the equation (7) for $A_{(t)}$ in equation (8), the internal dose was estimated for the liver and the bone of Mr. Kuboyama. The results of the estimation are given in Figs.19 and 20, where two values of θ, $\theta=\frac{1}{4}$ day and $\theta=1$ day, are assumed as the time at which the maximum activity might have reached the critical organ.

(Fig.19)

(Fig.20)

As can be seen in these figures, with this method of estimation the integral internal dose of radiation of the liver may be estimated to be about 4.7×10^2 rads for $\theta=1$ day and 7.1×10^2 rads for $\theta=\frac{1}{4}$ day, assuming T=15 days, and about 4.7×10^4 rads for $\theta=1$ day and 7.6×10^4 rads, for $\theta=\frac{1}{4}$ day, assuming T=10 days, while that of the liver about 1.9 rads for $\theta=1$ day and 2.2 rads for $\theta=\frac{1}{4}$ day, assuming T=100 days and about 40 rads for $\theta=1$ day and 58 rads for $\theta=\frac{1}{4}$ day, assuming T=30 days.

Although the highest value of the initial maximum activity estimated for the liver with $\theta=\frac{1}{4}$ day seems to be somewhat overestimate, judging from the degree of strong local radioactive

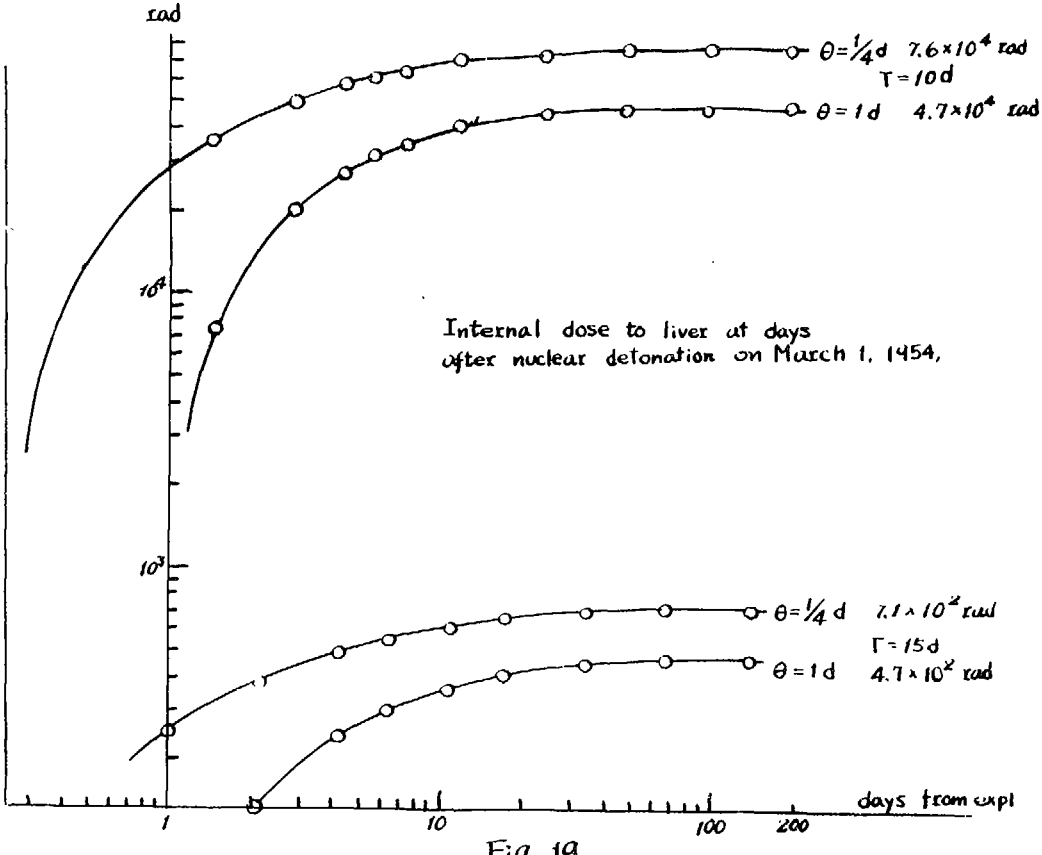
rad
10^4
10^3
θ=¼d 7.6×10⁴ rad
T=10d
θ=1d 4.7×10⁴ rad
Internal dose to liver at days
after nuclear detonation on March 1, 1954,
θ=¼d 7.1×10² rad
T=15d
θ=1d 4.7×10² rad
1
10
100
200
days from expl

Fig. 19

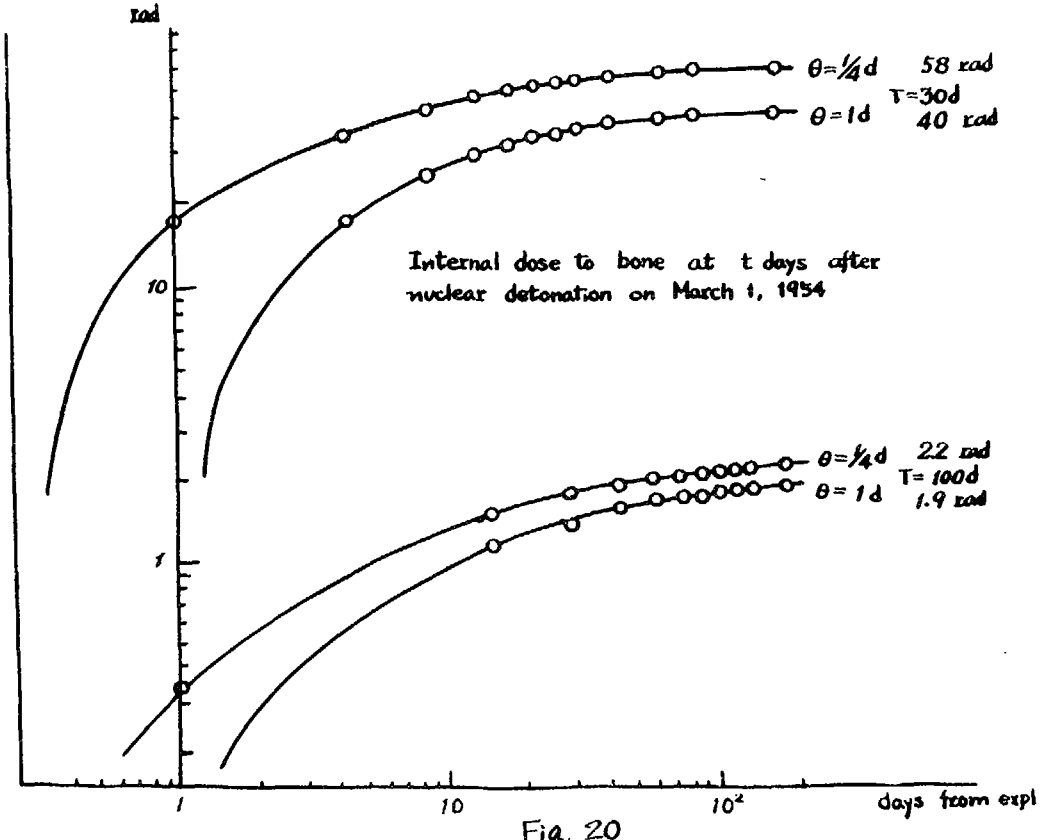
rad
θ=¼d 58 rad
T=30d
θ=1d 40 rad
Internal dose to bone at t days after nuclear detonation on March 1, 1954
10
1
θ=¼d 2.2 rad
T=100d
θ=1d 1.9 rad
1
10
10²
days from expl

Fig. 20

fallout and the conditions on the boat, it seems probable that the upper limit of the integral internal dose of radiation of some of the most highly exposed crew might have been in the range of the order of magnitude of 10^2 - 10^4 rads for the liver. However, since the individual differences in the function of liver would be quite large depending on various physiological and pathological conditions, the individual differences in the response of the liver to radiation may be expected to be considerably large depending on the various conditions of the individuals especially in the case of the abnormal functions of the liver.

In spite of the fact that a higher activity was detected in the bone at the time of Mr. Kuboyama's death, the initial maximum amount of radioactivity in the bone was estimated to be lower than that in the liver. This is because longer half-lives were assumed for the bone at the back extrapolation with the above method of estimation. However, from the comparison of the initial maximum amount of radioactivity in the bone with that in the liver, taking into consideration of the ratio of the initial retention of gross activity in the bone to that in the liver estimated from the biological experiment, the initial maximum amount of radioactivity estimated for the bone by the back extrapolation with the above method seems to be an underestimate. Although the corresponding integral internal dose of radiation for the bone is estimated in the range of about 2 - 60 rads as shown in the figure, if the initial amount of radioactivity were higher, the corresponding integral dose would be expected to be higher. Especially, if we take into consideration of the possible non-uniformity factor of 5 as given by ICRP(1958),[35] the integral dose for the bone might be estimated much higher. Judging from these considerations, it may not be too much to assume that the integral internal dose of radiation for the bone might have been in the range of the order of magnitude of about 10 - 300 rads.

7. Later Findings on the Contamination of Fish in 1954

On March 19th, 1954, the U.S. Atomic Energy Commission announced the enlargement of the warning area up to a semi-circle of radius about 450 miles extending north from the original Bikini-Eniwetok warning area. However, later the Bureau of Marine Products of the Japanese Government set a reporting area around Bikini which is more than double larger than the warning area expanded by the USAEC, and any fishing boats entering this area were requested to report to five major fishing ports in Japan for radiation monitoring.

The first contaminated fish which were brought back to Japan by No.5 Fukuryu Maru in the middle of March were found to be emitting much stronger radiation from the surface than from the inside, and the government set the tentative discarding level of radioactive contaminated fish at 100 cpm as measured at 10 cm from the wet surface with the beta-ray counter with 3.5 mg/cm^2 mica window plus 2.5 mg/cm^2 plastic cover to protect the counter window. The natural counts of this counter were about 30 cpm.

In the mean time the second thermonuclear explosion test was conducted on March 27th, the third on April 6th, the fourth of April 26th, the fifth on May 5th, in the South Pacific, and another test appeared to have been conducted sometime between May 8th - May 14th as judged from the analysis of the artificial radioactivity in the rain.

Although it was announced on May 14th, by the U.S.AEC that the series of H-bomb tests which were successfully conducted at Bikini area were concluded for that year, the contamination of fish and boats continued to be observed.

Therefore, in close cooperation with the public health officers of the Osaka City and Osaka Prefectural Government,

we have been continuing the radiation monitoring of the fish and the boats coming to Osaka Port from the South Pacific area.

From the fish which have been caught outside the expanded warning area, stronger radioactivity has been detected in the internal organs than on the skin. As the time went on, the area in which the contaminated fish were caught seemed to expand gradually.

In September, some of the fish caught at about 10 - 20 miles off the Pacific coast of Japan have been found to be contaminated (above 100 cpm at 10 cm), while no significant increase of radioactivity of ser-water near the coast of Japan has been detected at that time. Even in December highly contaminated fish (1000 - 2000 cpm) have been found occasionally among the fish caught in the coastal waters.

However the percentage of such highly contaminated fish seemed to be decreasing with time (0.1 - 0.01 per cent). Most of the fish were found to have the radioactive contamination of lesser degree, (apparent radioactivity of less than 100 cpm at 10 cm).

With most of these low contaminated fish the radioactive contamination of the white part of meat was estimated to be 10^{-4} μc/gm or less. The amount of fish consumed at a meal may be roughly about 100 - 300 gm.

The radioactive contamination of some of the fish contaminated to a medium extent examined by random sampling by the author and his associates at the Department of Biophysics, Osaka City University since May, may be estimated to be roughly about 10^{-1} - 10^{-2} μc/gm in the internal organs, 10^{-2} - 10^{-3} μc/gm in the red part of the meat, about 10^{-4} μc/gm in the white part of the meat, and less than 10^{-4} μc/gm in the skin.

Towards the end of 1954, besides the ordinary fission products, about 10 - 50 per cent of the apparent radioactivity detected in the fish has been found to be due to radioactive zinc Zn^{65}. The

reason why so much radioactive zinc accumulated in the fish is not certain, but the production of detectable amount of Zn^{65} by the neutron activation of some metallic part of the bomb itself seems to be possible at the time of the nuclear detonation.

At the end of the year the Japanese Government ordered the local governments to discontinue the radioactive examination of the fish partly because of the decreasing percentage of the highly contaminated fish, partly because of the larger maximum permissible amount of Zn^{65} given in the U.S.National Bureau of Standards Handbook 52,(36) and partly presumably because of the complicated economic problems accruing to the radiation monitoring at different ports scattered all over Japan. The total amount of fish discarded from March through December, 1954 because of the radioactive contamination (above 100 cpm at 10 cm) was estimated to be slightly over one million pounds.

(Fig.21)

According to the statistics in 1952,(30) the total amount of fish caught per year by the Japanese fishermen was about 9.6×10^9 pounds, and the total number of fishing boats was 446,652. Fig.21 shows the position of the boat operating in the Pacific when either the fish caught or the boat itself have been found to be contaminated by the radioactive dust on returning to the Japanese port during the period from March through June 1954.

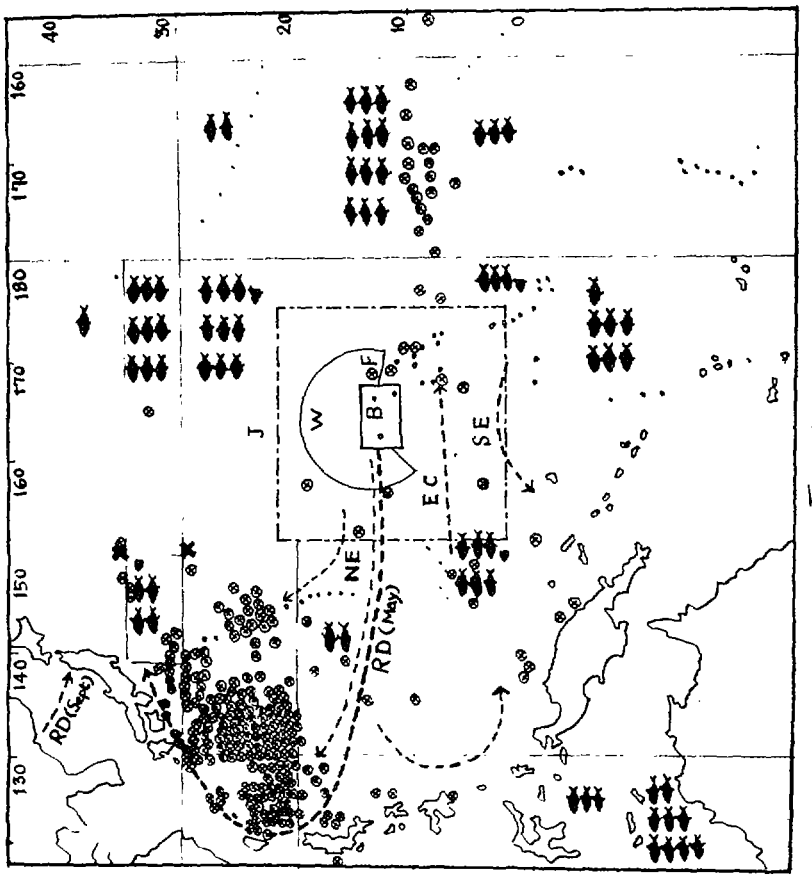
40
30
20
10
0
160
170
180
170
160
150
140
130
J
W
B
F
E C
SE
NE
RD(May)
RD(Sept)

Fig. 21

Fig. 21 : Radioactive Contamination of Boats and Fish in The Pacific in 1954 (III–VI)

B: Bikini, the small rectangle in solid line around Bikini indicates the original warning area on March 1st, 1954 (N 10°15' - 12°45', E 160°35' - 166°16').

⊗ F: Position of No. 5 Fukuryu Maru at 3.40 a.m. March 1st, 1954, which was identified to be N 11°53', E 166°58', about 80 - 90 miles east of Bikini.

⊗ Each cross with circle indicates the position of one boat operating in the Pacific when either the fish caught by the boat or the boat itself have been found to be contaminated by radioactive dust on returning to the Japanese port. (March through June, 1954).

W: The semi-circular area of radius 450 miles extending towards North from Bikini-Eniwetok Area indicates the expanded warning area during the period from March 19th to May 22nd, 1954. The remaining radioactive contamination of sea-water in this area detected by the Japanese Government Radiation Survey Boat in June may be estimated to be about $5 \times 10^{-2} - 10^{-2}$ μc/l. In August Rongerick and Rongelap Areas were announced to be still hazardously contaminated. (Notice to the Japanese Department of Hydrography from U.S.; Yomiuri, August 28th, 1954).

J: Large square in dot and dash line indicates the Reporting Area set by the Bureau of Marine Products of Japanese Government. (N 2° - 21°, E 152° - 175°). Any fishing boats which entered this area had been requested to report to five major fishing ports in Japan for radiation monitoring.

NE: North Equatorial Current.

EC: Equatorial Counter Current.

RD(May): Possible route along which the radioactive dust in the air has been carried to Japan during the period of unusually highly radioactive rainfall in the middle of May. (Kagaku, August 1954).

RD(Sept.): Possible direction of air current during the period of unusually highly radioactive rainfall in the middle of September. (Kagaku Asahi, December 1954).

: One fish indicates 10,000 fish per year caught by the Japanese fishing boats, in the area indicated. (Statistics from April 1952 to April 1953; Shizen, July 1954).

Summary

A study has been made, from the health physics standpoint, of the radioactivity emitted from the dust collected from No.5 Fukuryu Maru, which was showered by the strong radioactive ash at about 80 - 90 miles east of Bikini on March 1, 1954.

The probable dose of external gamma radiation the crew might have received during their two weeks voyage may be estimated roughly about 500 - 800 rad. However, judging from the strong radioactive contamination of the boat, it may be inferred that the crew might have received a considerable degree of internal irradiation besides the external whole body gamma irradiation and the local beta irradiation on the skin where the radioactive dust directly contacted. The specific activity of the dust when it fell on the boat a few hours after the nuclear detonation may be estimated to he roughly about one curie per gram.

From the radiochemical analysis and the beta-ray analysis, the major part of the radioactivity included in the dust was found to be due to a mixture of various fission products, while the main chemical component of the dust (Bikini ash) itself consisted of a calcium compound.

The alpha-ray was also detected by the use of an ionization chamber in that portion where transuranium elements, if present, ~~if present,~~ were collected.

The tuna fish and the shark fins which were brought back to Japan by No.5 Fukuryu Maru in the middle of March, 1954 were most strongly contaminated on the skin, but the fish caught later in the South Pacific were found to be contaminated more strongly in the internal organs rather than on the skin. The area in the Pacific where the radioactive contaminated fish were caught seemed to expand gradually with time. In the latter half of 1954, besides the ordinary fission products, the radio-

active Zn^{65} which was not found in appreciable amount in the original Bikini ash has been detected from the internal organs of the contaminated fish. A possible production of Zn^{65} by the neutron activation of some metallic part of the bomb devices at the time of nuclear detonation may be inferred.

Based upon the results of the biological experiments conducted with the extract of Bikini ash using rats and the result of the radioactive analysis of some of the organs of Mr. Kuboyama, the chief wireless operator of No.5 Fukuryu Maru who died at about 206 days after the nuclear detonation on March 1, 1954, an attempt has been made to estimate the internal dose of radiation. Although the accurate estimation seems to be extremely difficult, judging from the degree of strong radioactive fallout and the various conditions of the boat and the crew, the upper limit of the integral internal dose for the liver might be estimated in the range of the order of magnitude of about 10^2-10^4 rads, while that of the bone in the range of about 10 - 300. rads.

From the degree of strong radioactive contamination of the boat and the results of the radiochemical analysis of the Bikini ash, the size of the nuclear detonation conducted on March 1, 1954 in Bikini was estimated to be roughly about the order of magnitude of 10 megaton TNT equivalent fast neutron fission of the natural uranium or uranium 238.

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Studies on the Radioactive Contamination due to Nuclear Detonations II

(Preliminary Findings on the Radioactive Fallout due to Nuclear Detonations)

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Preliminary Findings on the Radioactive Fallout due to Nuclear Detonations

Since we have detected a considerable amount of artificial radioactivity in the rain in spring 1954, it has become one of the most important items, from the health physics point of view, to continue measurements of radioactivity in the rain and in the atmosphere. To watch out the radioactive contamination of our environment due to repeated nuclear weapons testings in other countries was also considered to be important from the nuclear engineering point of view, in the sense that the permissible allowances of the radioactivity for the peaceful uses of atomic energy might be lowered if the degree of radioactive contamination due to nuclear testings should continue to increase gradually and indefinitely.

If the permissible level were lowered, the cost for radiation protection may be expected to increase at the peaceful uses of atomic energy and should the radioactive contamination increase seriously in the future, it was anticipated that we may have to face a very difficult situation in designing the atomic energy facilities for peaceful purposes in our country.

From these points of views, we have been continuing measurements of the radioactivity in the rain in Osaka Japan since the spring of 1954. Some of the preliminary findings are introduced in this paper.

I Preliminary Findings[1][7][8]

Since a series of thermonuclear tests in the Pacific area began in March 1954, we have been recognizing an increase of radioactivity in the dust included in the rain.

Early March (March 6), 1954, a fallout of strange dust with the rain was observed over Atsumi Peninsula, central part of Japan mainland, facing the Pacific Ocean. Same phenomena were observed also in Yoshino district of Nara Prefecture near Osaka (March 6 -11). Later this dust was confirmed to be radioactive.

Since it was confirmed in our laboratory that the radioactivity detected in the rainwater which fell in Osaka on April 17 - 18 was mainly due to the artificial radioactive nuclides (about 50% of beta-ray activity was found to be due to rare-earth elements mixture) by the radiochemical analysis as well as by the analysis of decay curves of gross activity (apparent half-life was about one week), we have started a systematic checking of all rainfalls in Osaka. In Fig.1, the absorption curve of the radiation emitted from the radioactive residuals which were obtained from the rainwater of April 17 was compared with that of the radioactive dust which fell over the Atsumi Peninsula and the Yoshino district in the middle of March.

(Fig.1)

Since such naturally occurring radioactive substances as the disintegration products of Radon (Rn) or Thoron (Tn) might be included in the rain, we have carefully examined about this point. From the rapid measurement of the atmospheric dust, the rapid decay of RaB with the falf-life of about 27 min. and of RaC with the half-life of about 20 min. may be observed, but in a few hours RaB+RaC decays out to a negligible level. A few hours after sampling of the atmospheric dust the activity of ThB with the half-life of about 10.6 hrs may be observed but it decays out to an almost negligible level in

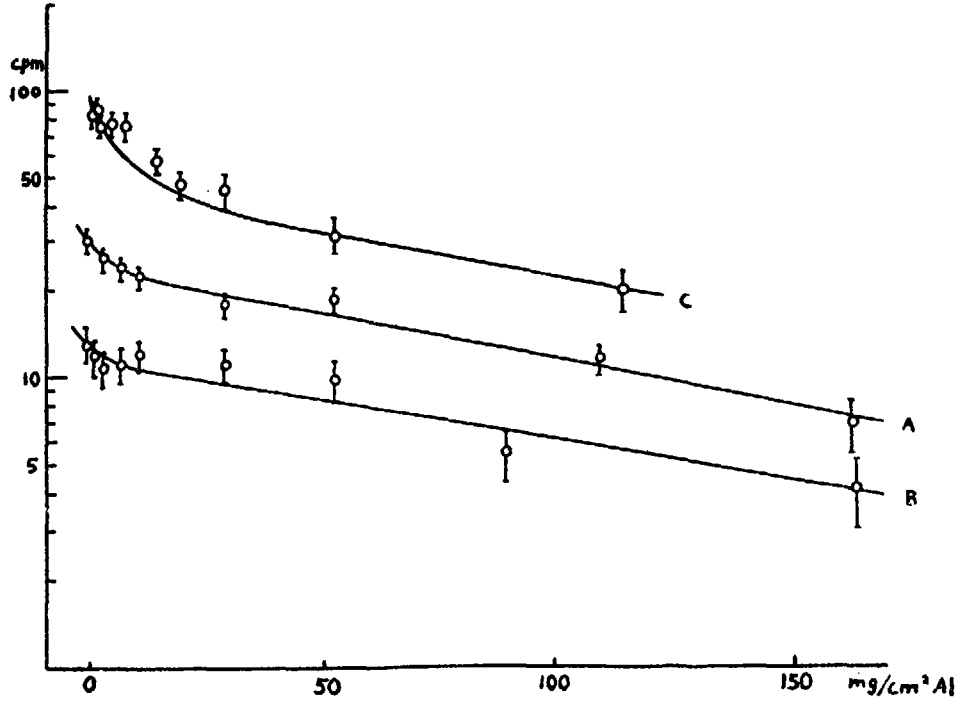


Fig. 1. The absorption curves of the radioactive dusts which fell in the middle of March over Atsumi Peninsula, Aichi Prefecture (A) and Yoshino district of Nara Prefecture (B) near Osaka are compared with that of the residual of April 17th rain in Osaka (C).

a day or so and in a few days it is almost undetectable with our usual method of routine measurements of the radioactivity of the rain.

Although the atmospheric content of such naturally occurring disintegration products of Rn or Tn may be expected to change considerably at different places, at different seasons of the year and at different times of the day, the percentage of the activity of ThB to that of RaB+RaC as estimated by back-extrapolation may be estimated to be about a few percent in the city of Osaka.

Since it takes a considerable time before measurement of radioactivity after sampling in order to collect the dust by complate evaporation of rainwater with our usual method of routine measurements, the possible degree of complication due to natural radioactivity such as Rn or Tn was considered to be almost negligible if we estimate the artificial radioactivity at the time of sampling by the back-extrapolation from the decay measurements a few days after sampling, especially when the artificial radioactivity due to nuclear testings included in the rain in Japan which is usually at least a few days to about a week of age is strong.

The measurement of radioactivity of the rainwater was carried out with the residuals, which were obtained by complete evaporation of the rainwater at least six to ten hours after sampling and was placed at a few centimeters from the beta-ray counter with 1.9 mg/cm^2 mica window and effective area π cm^2. The natural count of this type of counter is about 20 cpm and it is customary to express the radioactivity with the number of counts per minute detected above the natural background activity as estrapolated back to the time of sampling based upon the decay measurements for a few days at least.

In our past experiences, non-metalic container seemed better than metalic container for collection of rainwater for the purpose of examining radioactivity, so we have been using glass containers. Later it was found that the polyvinyl or polyethylene container or sheet might be more suitable for this purpose than the glass wares.

The radioactivity in units of microcuries was estimated from the comparison with the sub-standard source of $Sr^{90} - Y^{90}$. However, Osaka being a large industrial centre, the amount of residual after complete evaporation of rainwater containing soot and dust is sometimes so high that the self absorption due to thickness of sample at the time of radioactivity measurement may not be negligible. The apparent total radioactivity was occasionally found to be higher after the chemical separation of the radioactive subatances from the non-radioactive residuals.

The radioactivity detected in the rain in the middle of May 1954, was unusually high in Osaka as was in other districts.[1][2][4] From the initial rainfall on May 16, 1954, the radioactivity of about 87,000 cpm per litre was detected in Kyoto near Osaka by Prof. Shidei of Kyoto University[1][2][4] which may be estimated to correspond to about 0.5 - 1.0 micro-curie per litre. The radioactivity concentration of the rainwater was observed to be much higher at the beginning of rainfall. The amount of radioactivity washed down with the rain during this period of unusually highly radioactive rainfall may be estimated to correspond to roughly about 0.1 - 0.3 micro-curie per sq. m per day in Osaka.

In Fig.2, the absorption curves of the radioactivity detected in the rain on May 14 and on May 17 are shown.

(Fig.2)

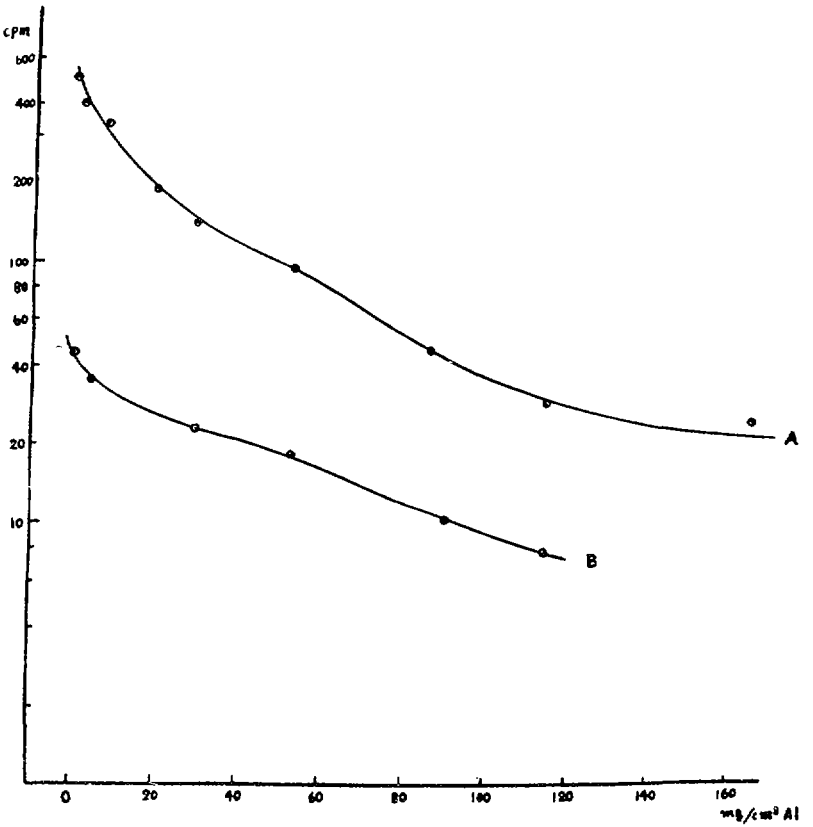


Fig. 2 The absorption curve of the radioactive residuals of May 17th rain (A) is compared with that of May 11th rain. (B)

The decay curves of gross activity detected in the rain of May 14 and May 17 are compared in semilogarithmic plots in Fig.3.

(Fig.3)

However, as can be seen in Fig.3, the decay of the gross activity of May 17 rain is faster than that of May 14 rain in Osaka which is due to the explosion on May 5, so we may have to assume that the unusual increase of radioactivity in May 17 rain might be due to some other tests conducted after May 5 which has not been ascertained from the barograph and microbarograph records.

If it could be assumed that the decay curve of gross activity of radioactive contamination of rainwater should be linear in logarithmic plot when the time of origin is taken as the time of nuclear explosion, the possible date of explosion after May 5 may be estimated to be about May 12 or sometime between May 8 and 14, 1954.

However, since the nuclear tests had been conducted repeatedly in the South Pacific in the period from March 1 to May 13, 1954, it may be possible that in the rainfall during May the radioactive dust originated in different tests were mixed up to some extent. Under such circumstances a more accurate estimation of the probable date of explosion from the analysis of the decay curve of gross activity in the rain may become rather difficult.

On April 19, 1955, an unusually thick whitish rain was observed to have fallen over a considerably wide area (Osaka, Kobe, Nara districts and a part of Kyoto Prefecture). Usually the residual obtained after complete evaporation of rainwater was black or dark brownish in Osaka because of a great percentage of soot.

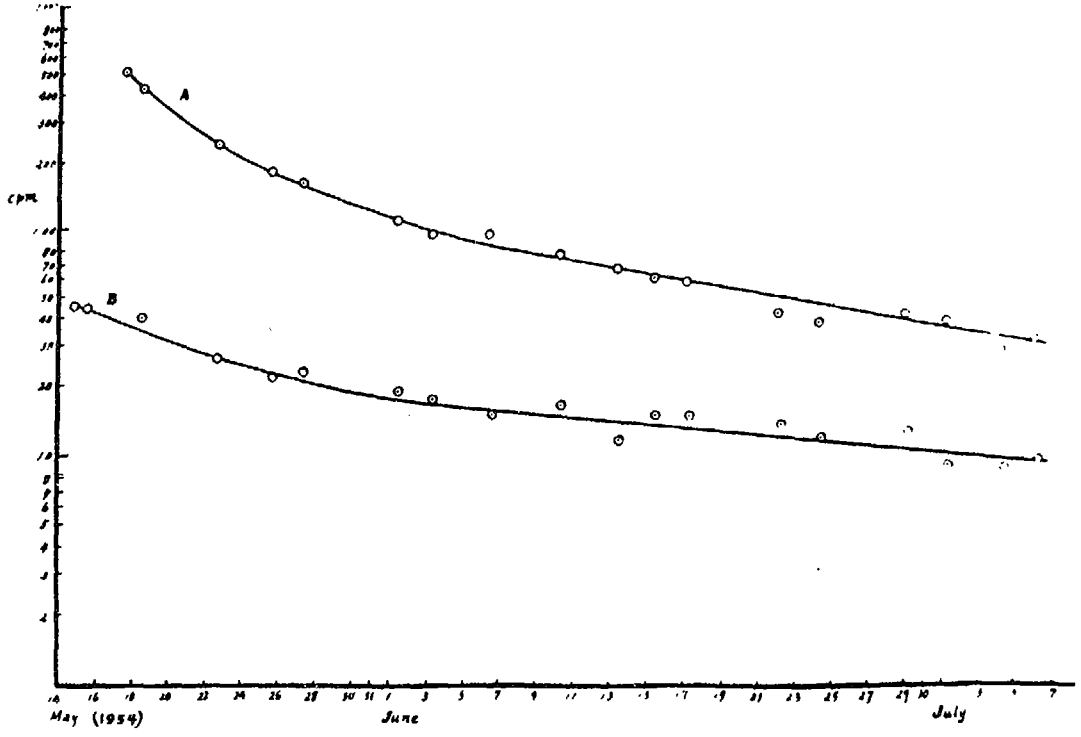


Fig. 3 The decay curve of the radioactive residual of May 17 rain (A) is compared with that of May 14 rain (B) in semilogarithmic plot.

Although the amount of rainfall was only about 0.1 - 0.2 mm in Osaka on April 19, it was so thick that each drop made a clear white spot on the roof, window panes, and motor-cars. From the rainwater collected in the glass container 2,600 - 3,500 cpm per litre of radioactivity were detected, while the apparent radioactivity detected from the rainwater collected in the polyvinyl container was found to be only about one-tenth of the above figure, in spite of the fact that the polyvinyl vessel is considered to be better than the glass container as the rainwater collector for the radioactivity measurement. Most of the thick whitish substance in the rain including radioactive substances seemed to have adhered to the polyvinyl membrane. In case of the unusually thick rain we may have to pay special attention to the method of collecting rainwater and of transferring it to other vessels from the original collector. Otherwise there may be a great danger of losing a greater part of radioactive dust in the rain before the radioactivity measurement with the residual is done.

From the quantitative chemical analysis of April 19 rain, the whitish residual was found to contain over 50% calcium. From the analysis of the dust collected from a large funnel used for rainwater collection on April 19 at the Osaka Central Meteorological Station, it was found that the dust contained about 30% calcium, 40% silica, and the rest was composed of soot and other dust. From further spectroscopic analysis of the dust by emission spectrum conducted in cooperation with Mr. Shimizu of Osaka City Technical Research Institute, the following elements were confirmed: Ca (a large quantity), Si, Fe, Al, Mg, Na, a minute trace of Ni and B. The dust in the

rainwater in the middle of March had been found to contain only about 5 - 6% calcium, about 40% silica and over 50% soot and other dust.

From the preliminary radiochemical analysis of April 19th dust with ion exchange method (Dowex 50) conducted in cooperation with Dr. Yamadera of the department of inorganic chemistry, the radioactivity remaining after about a week was confirmed to be due mainly to the various fission products including rare-earth elements which amounted to about 60 to 70% of the total beta-ray activity. However, the calcium and silica included in the residual did not seem to be radioactive to an appreciable extent.

Judging from these findings it seems probable that the minute particles of calcium and silica might have absorbed some of the radioactive dust in the atmosphere because of their high surface activity, and were washed down with the rain in addition to the radioactive dust directly collected by the rain. However, from where such high content of calcium in April 19 rain came is still unknown. On April 13, similar rainfall was reported to have been observed in Hokkaido, the northernmost island of Japan, and later in some districts of the western part of Japan mainland including Osaka. The possible corelation with the yellow sands from continental China was also considered.

In the middle of September and early November, 1954, highly radioactive rainfall was reported mostly in the northern part of the Japan mainland.[3] On Sept. 22, the radioactivity of 128,000 cpm per liter with the apparent shorter half-life was detected in Yamagata Prefecture by Prof. Terasaki of Yamagata University,[3][5] which was estimated to correspond to about 0.3 microcurie per litre with the substandard source of Sr^{90} - Y^{90}.

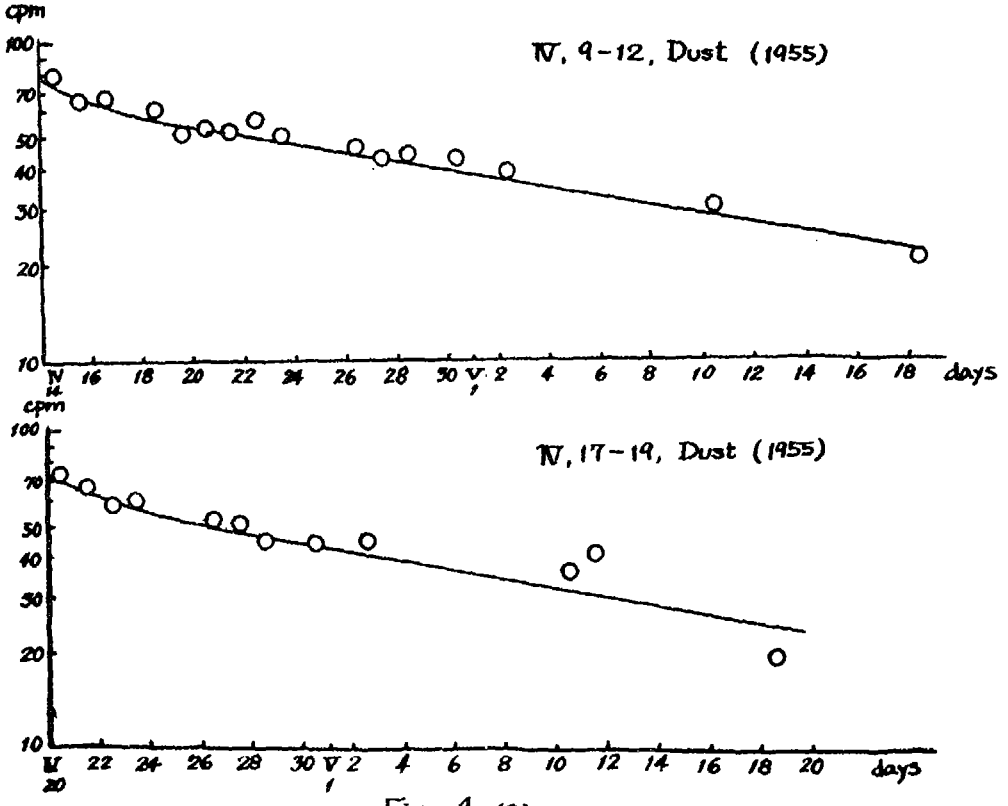
In Osaka, although a slight increase of radioactivity in the rain was observed a few days later, no such high increase of radioactivity in the rain as reported in the northern part was observed during this period.

The occasional increase of radioactivity in the rain since February 1955 may be considered to be due at least partly to the effects of the repeated atomic tests conducted in Nevada atomic testing ground since early 1955.

Some of the other examples of the decay measurements of the radioactivity of the dust samples collected in Osaka in spring of 1955 are given in Fig.4.

(Fig.4)

In order to check whether such a high degree of radioactive fallout as observed in the rain and dust of Osaka, Japan in 1954 would be possible, we have examined an order of magnitude estimation of the global fallout assuming a fission-fusion-fission type of nuclear detonation.


cpm
100
70
50
40
30
20
10
IV, 9-12, Dust (1955)
IV 14 16 18 20 22 24 26 28 30 V 2 4 6 8 10 12 14 16 18 days
cpm
100
70
50
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20
10
IV, 17-19, Dust (1955)
IV 20 22 24 26 28 30 V 2 4 6 8 10 12 14 16 18 20 days

Fig. 4 (a)

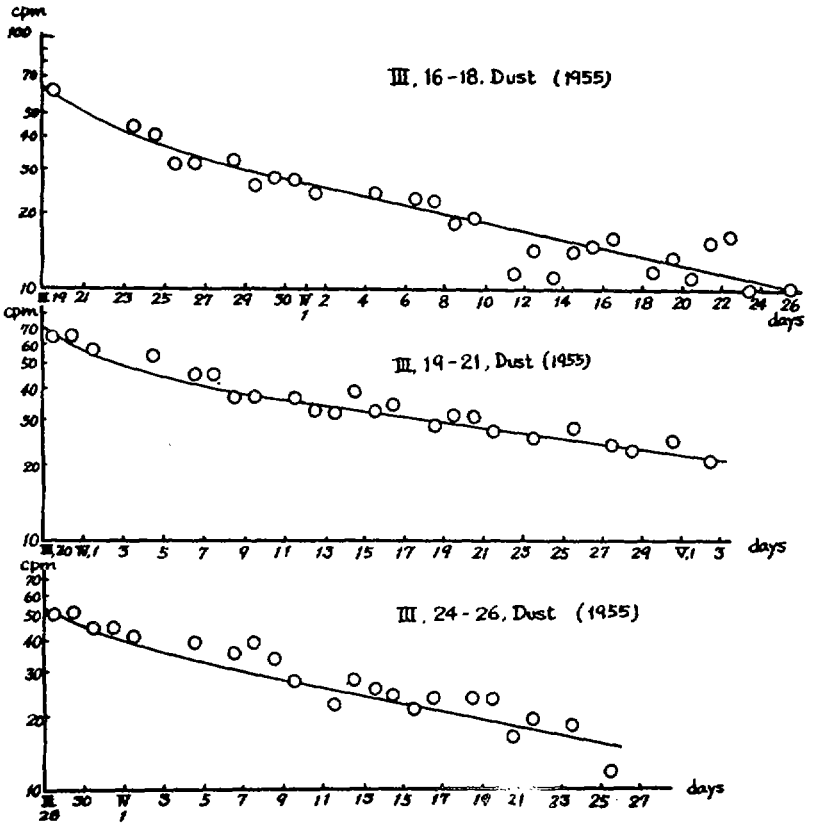

cpm
100
70
50
40
30
20
10
III. 16-18. Dust (1955)
III. 19 21 23 25 27 29 30 IV 2 4 6 8 10 12 14 16 18 20 22 24 26
1
days
cpm
70
60
50
40
30
20
10
III, 19-21, Dust (1955)
III, 20 IV, 1 3 5 7 9 11 13 15 17 19 21 23 25 27 29 V, 1 3
days
cpm
70
60
50
40
30
20
10
III. 24-26, Dust (1955)
III 28 30 IV 1 3 5 7 9 11 13 15 17 19 21 23 25 27
days

Fig. 4 (b)

II Order of Magnitude Estimation

1. Fission-Fusion-Fission Bomb[9][10]

The total energy of about 200 Mev is assumed to be released per one fission of uranium-235, plutonium-239 or uranium-238 corresponding to a reduction of mass by about one tenth of a percent. Of the total energy, the kinetic energy of the prompt neutron emitted during fission is about 2.5% and the number of delayed neutrons emitted during the period from a fraction of a second to a few seconds after fission is about a few percent of the prompt neutronts or less. The energy of prompt gamma rays is about 2.3%. The energy of gamma rays emitted from the fission products is about 2.9%, while that of beta rays about 3.4%. The energy of neutrino which may not be available as heat energy may be estimated to be about 5.5%. Therefore the energy released as the explosion energy at the time of fission may be estimated to be about 80 - 90% of the total fission energy.

Based upon the above figure, the energy release by the fission of 1000 Kg fissile material which corresponds to about 2.6×10^{27} fissions may be estimated to be 8.4×10^{23} ergs or 2×10^{16} g. cal. Since 1 Kw.hr = 860 Kcal, this may correspond to about 2.3×10^{10} Kw.hr, or assuming the energy release of 1 g TNT explosion is about 1 Kcal, the nuclear fission of 1000 Kg fissile material may be estimated to correspond to about the explosion of 20 megaton TNT.

Since such a large amount of fission seems to be almost impossible with the ordinary atomic bomb of Hiroshima or Nagasaki type, the nuclear bomb of Bikini type is assumed to be 3F-bomb or fission-fusion-fission type. It is assumed that the thermonuclear reaction or fusion reaction using lithium-6 deuteride was first ignited by the slow neutron fission of U^{235} or Pu^{239} and then a

large amount of uranium 238 used as a tamper might be fissioned by the fast neutrons released by the fusion reaction according to the following scheme.

$$\left.\begin{array}{l}\text{(fission of } Pu^{239}) \Rightarrow {}_0n^1 + {}_3Li^6 \rightarrow {}_1T^3 + {}_2He^4 + 4.8\ \text{MeV} \\ {}_1D^2 + {}_1T^3 \rightarrow {}_2He^4 + {}_0n^1 + 17.3\ \text{MeV} \\ {}_1D^2 + {}_1D^2 \begin{cases} \nearrow {}_2He^3 + {}_0n^1 + 3.2\ \text{MeV} \\ \searrow {}_1H^1 + {}_1T^3 + 4.0\ \text{MeV} \end{cases}\end{array}\right\} \Rightarrow (\text{fission of } U^{238}) \quad (1)$$

At the first step of slow neutron fission, about 25 Kg of plutonium may be necessary. Usually a fraction of this amount may be assumed to fission initially, but even if we assume 25 Kg of plutonium fissioned because of the high neutron flux released by the second step, the corresponding energy release may be estimated to be equivalent to about 0.5 megatons of TNT. However, since most of the neutrons from the fission of plutonium may not have sufficient energy to cause the fission of U^{238}, the main role of fusion reaction using $L_i{}^6D$ may be to produce fast neutrons to cause the fission of U^{238}. To release the explosion energy equivalent to 20 megatons of TNT, roughly about 1000 Kg of U^{238} may be necessary from the above estimation. To produce sufficient fast neutrons for this purpose, about 30 Kg of $L_i{}^6D$ may be necessary and the corresponding energy release from this reaction may be estimated to be equivalent to roughly about the order of 3 megatons of TNT. From these considerations roughly about 80% of the energy released by the above type of bomb may be estimated to be derived from the fast neutron fission of uranium-238.

Although it has been reported[10] that most of the nuclear detonations of megaton size conducted by the United States in the past were about 50% fission and 50% fusion, if we assume that the fission of 1000 Kg U^{238} corresponds to the nuclear detonation equivalent to 20 megatons of TNT, the corresponding number of fissions would be about 2.6×10^{27}. Therefore, assuming the rate of neutron release of about 2.5 per fission, the number of neutrons generated per 20 megatons would be about 6.5×10^{27}. Dividing this number by the Avogadro number, we may estimate the corresponding total mass of neutrons to be about 10.8 Kg.

2. Carbon-14

In case of the nuclear detonation in the air, most of the neutrons released would be expected to react with the nitrogen nuclei in the air, producing the radioactive carbon-14 with the half-line of 5,600 years.

$$ {}_7N^{14} + {}_0n^1 \rightarrow {}_6C^{14} + {}_1p^1 + 0.61 \text{ Mev} $$
$$ {}_6C^{14} \rightarrow {}_7N^{14} + \beta^- + 0.16 \text{ Mev} \qquad (2) $$

Although the actual number of neutrons escaping into the air may depend on the type of nuclear detonation, if we assume roughly about 50% of the generated neutrons would escape into the air, about 74 Kg of radioactive carbon-14 may be produced. If we assume all of the neutrons estimated in the above would be used for this reaction, about 148 Kg or 672,000 curies of radioactive carbon may be produced per 20 megaton TNT equivalent nuclear detonation. If we assume all fusion reaction using only deuterium-tritium fusion reaction, roughly about 5 times more neutrons may be estimated to generate for the same energy release as compared with the above estimation, and therefore about 5 times more radioactive carbon-14 would be produced, if most of the neutrons should escape into the air. However, if we assume all fusion reaction of a mixed type

as shown in equation (1), the upper limit for the induced activity may be about ten times higher than the above estimation.

The amount of carbon-14 produced per year by the absorption of cosmic ray neutrons in the atmospere is estimated to be about 7-10 Kg per year. The natural carbon-14 content of the biosphere is estimated to be about 1.28 -1.46x10^{-12} times the total carbon content. The amount of naturally existing carbon-14 in the total earth reservoir is estimated to be about 56 to 81 metric tons with the distribution about 1.69 - 1.47% in the atmospheric carbon dioxide, about 3.88 - 2.83% in the terrestrial living matter and humus, about 6.7 - 8.61% in the total organic matter of the ocean, about 87.8 - 87.2% in the total inorganic matter of the ocean.

Assuming that the DNA in each diploid cell contains about 1.2x10^{11} atoms of carbon, the natural carbon-14 content in DNA may be estimated to be about 0.16 atom per cell with the disintegration rate per year about 2x10^{-5} . If we assume that the generative cells, on the average, would suffer 30 years of this kind of transmutation effect, the probability per generation per person may be estimated at about 6x10^{-4}.

Leipunsky,[11] Pauling[12] and Totter, et al[13] have estimated the possible biological hazard to man due to the transmutation effect of carbon-14 from nuclear weapons testing. Since the ratio of mutation over transmutation of carbon-14 in DNA is not exactly known with man, the exact estimation of the genetic hazard due to transmutation effect seems to be difficult.

However, if we assume the ratio of mutation over transmutation to be unity, the transmutation effect may not be considered negligible, although the addition of 148 Kg of carbon-14 corresponding to 20 megaton TNT equivalent nuclear detonation estimated in the above would mean only an addition of roughly about 0.2% to the total carbon-14 reservoir.

3, Gross activity of Fission Products

The nuclear fission of uranium or plutonium is known to take place in more than 40 different ways producing over 80 different nuclides. The fission yield curve against mass number is slightly displaced towards higher mass number for uranium-238 than for uranium-235. The primary nuclides, containing neutrons in excess usually, undergo a few beta decays sometimes accompanied by gamma rays to become stable nuclides.

Therefore, in the fission products, about 100 different radioactive nuclides with greatly different half-lives are mixed. Although the decay rate for such a complex mixture may be obtained as the sum of the exponential decay rates for each radioactive nuclides, the composite decay rate is known, according to Way and Wigner,[14] to follow approximately the following simple law.

$$I = A_o t^{-\alpha} \qquad (3)$$

where t is the time after nuclear fission, A_o a constant and the theoretical value of α is 1.2.

According to Hunter and Ballou,[15] the value of α for the fission products of uranium 235 may change somewhat with the time after fission as shown in the following.

$$A_{(t)} = 2.8 \times 10^{3} t^{-0.89} (1 \text{ min} \leq t < 30 \text{ min}) \qquad (4)$$

$$A_{(t)} = 5.7 \times 10^{3} t^{-1.11} (30 \text{ min} \leq t < 1 \text{ day}) \qquad (5)$$

$$A_{(t)} = 1.56 \times 10^{4} t^{-1.25} (1 \text{ day} \leq t < 4 \text{ days}) \qquad (6)$$

$$A_{(t)} = 2.4 \times 10^{3} t^{-1.03} (4 \text{ days} \leq t < 100 \text{ days}) \qquad (7)$$

$$A_{(t)} = 2.16 \times 10^{6} t^{-1.6} (100 \text{ days} \leq t < 3 \text{ yr}) \qquad (8)$$

where $A_{(t)}$ is the number of disintegrations per minute (dpm) of the fission products per 10^4 fissions at t=0 and t is the time in minutes after instant of fission. The actual total activity at t=0 is computed to be 4.4×10^4 dpm. Judging from this figure, 2.6×10^{27} fissions may be estimated to correspond to about 5.2×10^{15} curies at t=0.

However, for the approximate estimation, the rate of emission of gamma ray photons I_γ and that of beta particles I_β per second per fission may be expressed by the following equations.

$$I_\gamma = 1.6t^{-1.2} \qquad (9)$$

$$I_\beta = 3.2t^{-1.2} \qquad (10)$$

where t is the time in seconds after fission and the above expressions may be approximately valid from about 10 seconds after fission with the average limit of error less than a fator of about two or three in most cases.

If we express the time t after fission in unit of minutes and I_γ and I_β in unit of dpm, we may have the following expressions per 10^4 fissions.

$$I_\gamma = 7 \times 10^3 t^{-1.2} \qquad (11)$$

$$I_\beta = 14 \times 10^3 t^{-1.2} \qquad (12)$$

The comparison with the values estimated by Way and Wigner and the curves given by Hunter and Ballau[15] for 10^4 fissions of U^{235} is shown in Fig.5.

(Fig.5)

The average energy of beta rays is about 0.4 Mev and that of gamma 0.7Mev. The rate of emission energy in unit of Mev per second per fission for beta and gamma rays may be given on a rough approximation by the following expressions.

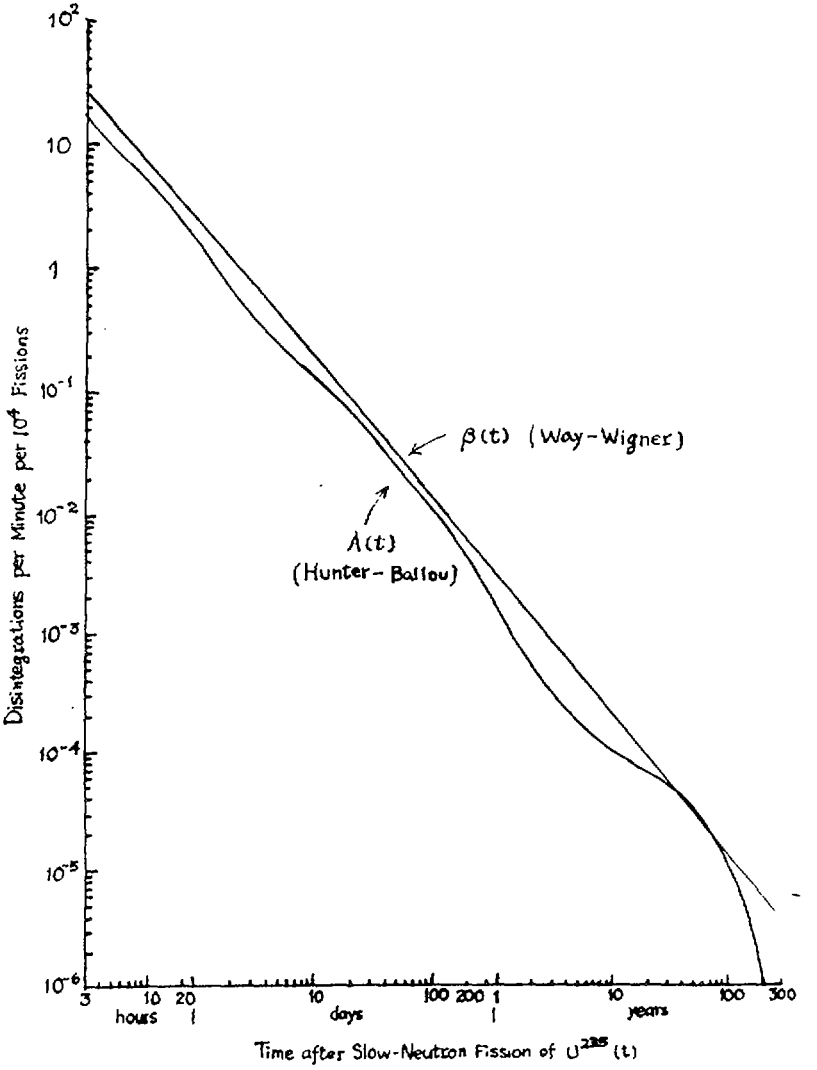
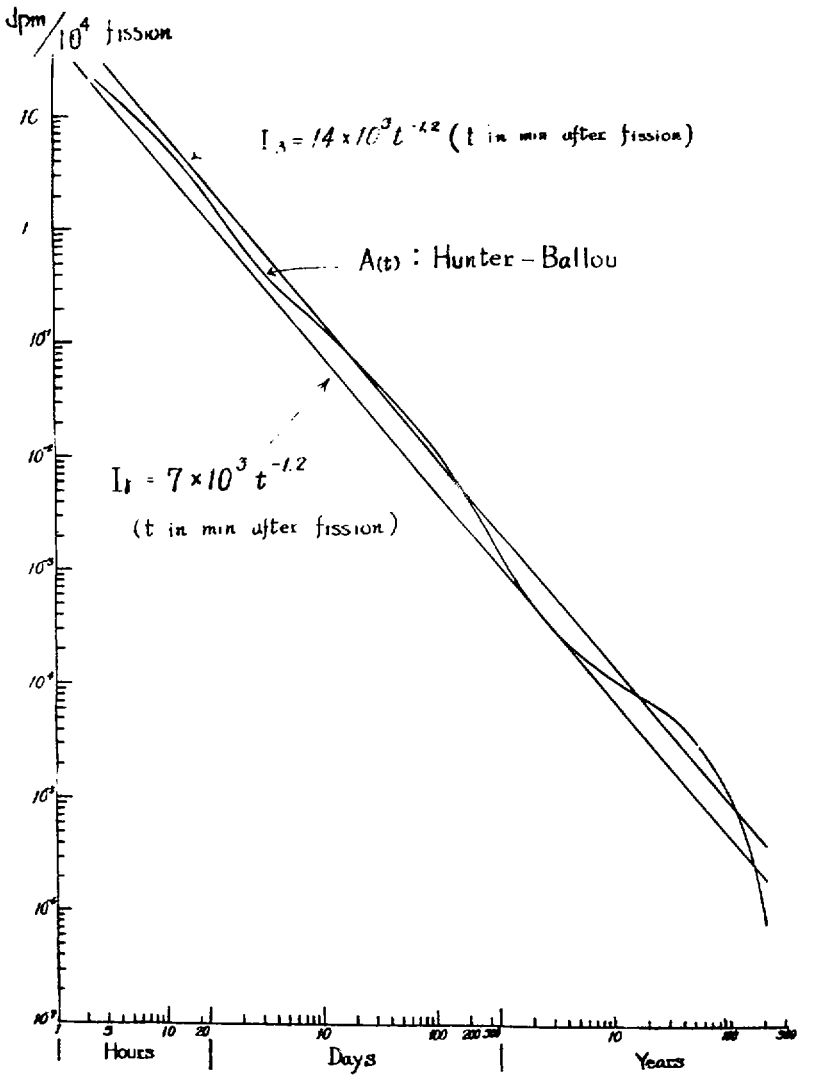

10^2
10
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
Disintegrations per Minute per 10^4 Fissions
$\beta(t)$ (Way-Wigner)
$\lambda(t)$
(Hunter-Ballou)
3
10
20
hours
10
days
100
200
1
10
years
100
300
Time after Slow-Neutron Fission of U^{235} (t)

Fig. 5 (a)


dpm/10^4 fission
10
1
10^-1
10^-2
10^-3
10^-4
10^-5
10^-6
10^-7
I_3 = 14 x 10^3 t^-1.2 (t in min after fission)
A(t) : Hunter – Ballou
I_1 = 7 x 10^3 t^-1.2
(t in min after fission)
1
5
10
20
10
100
200
300
10
Hours
Days
Years

$$E_\beta = 1.3t^{-1.2} \quad (13)$$

$$E_\gamma = 1.1t^{-1.2} \quad (14)$$

For 1,000 Kg of fission products corresponding to 2.6×10^{27} fissions or 20 megatons TNT equivalent nuclear detonation, the total gamma and beta activities may be expressed by the following equations.

For gamma activity,

$$I_\gamma \text{ (dps)} = 4.2 \times 10^{27} t^{-1.2} \quad (15)$$

$$I_\gamma \text{ (curies)} = 1.1 \times 10^{17} t^{-1.2} \quad (16)$$

For beta activity,

$$I_\beta \text{ (dps)} = 8.4 \times 10^{27} t^{-1.2} \quad (17)$$

$$I_\beta \text{ (curies)} = 2.2 \times 10^{17} t^{-1.2} \quad (18)$$

where t is the time in seconds after fission, and the above equation may be approximately valid from about 10 seconds after fission.

For gamma activity,

$$I_\gamma \text{ (dpm)} = 13 \times 10^{24} t^{-1.2} \quad (19)$$

$$I_\gamma \text{ (curies)} = 6 \times 10^{12} t^{-1.2} \quad (20)$$

For beta activity,

$$I_\beta \text{ (dpm)} = 26 \times 10^{24} t^{-1.2} \quad (21)$$

$$I_\beta \text{ (curies)} = 12 \times 10^{12} t^{-1.2} \quad (22)$$

where t is the time in hours after fission.

If we express the time t after fission in unit of days,

$$I_\gamma = 1.3 \times 10^{11} t^{-1.2} \quad (23)$$

$$I_\beta = 2.6 \times 10^{11} t^{-1.2} \quad (24)$$

From the above equations, the total activity of the fission products due to the fission of about 1,000 Kg of uranium-238 may be estimated to be about 2.6×10^{10} curies at one week, 4.5×10^{9} curies at one month and 1.2×10^{9} curies at three months after the nuclear fission. If we assume that the total activity estimated in the above were distributed uniformly over the earth's surface of about 5×10^{14} m^2, the specific activity per unit area may be estimated to be about 5.1×10^{-5} c/m^2 at one week, about 8.8×10^{-6} c/m^2 at one month and about 2.3×10^{-6} c/m^2 at three months.

4. External Gamma Dose

The gamma ray activity of fission products at t hours after fission is given by the expression(20) to be $6 \times 10^{12} t^{-1.2}$ for 20 megaton equibalent nuclear detonation. If we assume the fission products were uniformly distributed over the earth's surface of about 5×10^{14} m^2, the specific gamma activity per unit area may be estimated to be about $0.012\ t^{-1.2}$ c/m^2. If we assume the gamma activity of fission products were uniformly deposited over a plane surface in the area large enough as compared with the mean free path of gamma ray photons with the specific gamma activity of one curie per square meter, the dose rate received at a height of about one meter above the surface may be estimated to be about 8.3 rad per hour. Therefore the dose rate for the global uniform deposition of fission products at t hours after 20 megaton equivalent fission may be given by $0.1 t^{-1.2}$ rad per hour. Using this relation, the total dose received from the time t_1 to t_2 after detonation may be estimated by the following equation.

$$D(t_1 \rightarrow t_2) = \int_{t_1}^{t_2} 0.1\ t^{-1.2}\, dt \qquad \text{rad} \tag{25}$$

or

$$D(t_1 \rightarrow t_2) = 0.5\,(t_1^{-0.2} - t_2^{-0.2}) \text{ rad} \tag{26}$$

The infinity dose from the time t_1 after fission may be estimated approximately by putting $t_2 \rightarrow \infty$ in the above equation.

$$D = 0.5 t_1^{-0.2} \tag{27}$$

If we assume in general the specific surface activity at the time t after fission to be expressed by

$$I = A_0 t^{-\alpha} \tag{28}$$

where A_0 and α are constants, the total dose received for an exposure from time t_1 to t_2 may be given by

$$D_{(t_1 \rightarrow t_2)} = \int_{t_1}^{t_2} k A_0 t^{-\alpha} \, dt \tag{29}$$

where k is the dose rate for the deposition of unit specific surface activity. If we assume k is also a constant, the rquation (29) may be expressed as

$$D_{(t_1 \rightarrow t_2)} = \frac{k A_0}{\alpha - 1} t_1^{1-\alpha} \left[1 - \left(\frac{t_1}{t_2} \right)^{\alpha - 1} \right] \tag{30}$$

The infinity dose from the time t_1 to the time t_2 which is very large compared to t_1 so that $(t_1/t_2)^{\alpha-1}$ $(\alpha>1)$ is small compared to unity may be given by

$$D = \frac{k A_0}{\alpha - 1} t_1^{1-\alpha} \tag{31}$$

In this case, if the specific surface activity $I_{(t_1)}$ at the time t_1 after detonation is known, A_0 in equation (31) may be given by

$$A_0 = I_{(t_1)} t_1^{\alpha} \tag{32}$$

Substituting A_0 of equation (32) into equation (30), we may obtain,

$$D(t_1 \rightarrow t_2) = \frac{k I(t_1)}{\alpha - 1} t_1 \left[1 - \left(\frac{t_1}{t_2} \right)^{\alpha - 1} \right] \qquad (33)$$

Therefore the infinity dose from the time t_1 to infity may be given by

$$D = \frac{k I(t_1)}{\alpha - 1} t_1 \qquad (34)$$

where k is the dose rate from unit specific surface activity and $I_{(t_1)}$ the specific surface activity at the time t_1 after nuclear detonation.

In case of the dose due to gamma rays from the uniform deposition of fission products with $\alpha = 1.2$, the dose rate k at a height of about one meter may be estimated to be about 8.3 rad per hour corresponding to the specific activity of 1 c/m^2. Therefore, substituting these values into equation (33), we may obtain,

$$D_{(t_1 \rightarrow t_2)} = 42 I t_1 \left[1 - \left(\frac{t_1}{t_2} \right)^{0.2} \right] \qquad (35)$$

where I is the specific gamma activity (c/m^2) at the time t_1 after fission and t_1 and t_2 are expressed in unit of hours after nuclear detonation. The infinity dose D of the equation (35) may be expressed by the following equation in this case.

$$D = 42 (I \cdot t_1) \text{ rad} \qquad (36)$$

where D is the infinity dose from the time t_1 hours after fission and I the specific gamma activity per unit area expressed in unit of c/m^2 at the time t_1.

Using the above relations and assuming the gamma activity to

be roughly about one half of the beta activity, the infinity gamma dose may be estimated roughly about $\overset{at}{\Lambda}$180 mrad at 1 week, 131 mrad at 1 month and 105 mrad at 3 months for the uniform deposition of 20 megaton TNT equivalent fission products over the earth's surface, while the 30 year dose roughly about at 140 mrad at 1 week, 90 mrad at 1 month and 64 mrad at 3 months after the nuclear detonation.

According to R.E.Lapp,[16] the principal contributors to gamma dosage within the first year are 35d Nb^{95}, 65d Zr^{95}, 40d Ru^{103}, and 33d Ce^{141} with 1 yr Ru^{106} and 285d Ce^{144} predominating thereafter. Ten years after the detonation the gamma activity will be dominated by 28yr Cs^{137}. Therefore, in order to obtain more accurate estimates of the gamma dosage for t_1 longer than about a year, the estimation should be done based upon more accurate results of the radiochemical analysis of the fallout, but an order of magnitude estimation may be possible with the above method especially for a relatively small value of t_1.

5. Rate of Fallout

The rate of deposition per unit area per day (R) may be approximated by

$$R = kq \frac{I_{(t)}}{A} e^{-kt}, \qquad k = \frac{0.693}{T_a} \qquad (37)$$

where the total activity $I_{(t)}$ at t days after fission is assumed to be uniformly spread over the area A, and k is the fraction deposited on the ground per day, T_a the half elimination time in days from the atmosphere and q the fraction of total activity injected into the atmosphere.

The value of ke^{-kt} may be estimated to be 9.5×10^{-4} at one week, 9.2×10^{-4} at one month and 8.7×10^{-4} at three months for the stratospheric fallout with the half elimination time of about two years, and 1.95×10^{-2} at one week, 1.15×10^{-2} at one month, and 2.6×10^{-3} at three months for the tropospheric fallout with the half elimination time of about one month. Assuming the expression (24) for $I_{(t)}$ in equation (37), the rate of deposition of beta activity

of gross fission products per unit area per day may be estimated to be as follows.

Assuming q = 1/2 and A = 5×10^{14} m^2 for stratospheric fallout, At 1 week,

$$R_\beta = 2.4 \times 10^{-8} \text{ c/m}^2 - \text{day} \quad (38)$$

or

$$R_\beta = 5.4 \times 10^{4} \text{ dpm/m}^2 - \text{day} \quad (39)$$

At 1 month,

$$R_\beta = 4 \times 10^{-9} \text{ c/m}^2 - \text{day} \quad (40)$$

or

$$R_\beta = 8.9 \times 10^{3} \text{ dpm/m}^2 - \text{day} \quad (41)$$

At 3 months,

$$R_\beta = 1 \times 10^{-9} \text{ c/m}^2 - \text{day} \quad (42)$$

or

$$R_\beta = 2.2 \times 10^{3} \text{ dpm/m}^2 - \text{day} \quad (43)$$

Assuming q = 1/4 and A = 2.5×10^{14} m^2 the total area of Northern Hemisphere for tropospheric fallout,

At 1 week,

$$R_\beta = 5 \times 10^{-7} \text{ c/m}^2 - \text{day} \quad (44)$$

or

$$R_\beta = 1.1 \times 10^{6} \text{ dpm/m}^2 - \text{day} \quad (45)$$

At 1 month,

$$R_\beta = 5.2 \times 10^{-8} \text{ c/m}^2 - \text{day} \quad (46)$$

or

$$R_\beta = 1.2 \times 10^{5} \text{ dpm/m}^2 - \text{day} \quad (47)$$

At 3 months,

$$R_\beta = 3.2 \times 10^{-9} \text{ c/m}^2 - \text{day} \quad (48)$$

or

$$R_\beta = 7.1 \times 10^{3} \text{ dpm/m}^2 - \text{day} \quad (49)$$

Assuming 10% for the counting efficiency of beta ray and about 10 mm of rain fall, we may obtain the following order of megnitude estimates of the beta activity of the gross fission products corresponding to about 20 megaton equivalent fission of about 1,000 Kg of U^{238}. About 1.2×10^5 cpm/m^2 - day or roughly about 10^4 cpm/l at one week, about 1.3×10^4 cpm/m^2 - day or roughly about 10^3 cpm/l at one month, and about 9.3×10^2 cpm/m^2 - day or roughly about 10^2 cpm/l at three months after fission.

In case of the 20 kiloton equivalent slow neutron fission of about 1 Kg of U^{235} or Pu^{239}, if we assume that all the activity were uniformly distributed over the Northern Hemisphere with the area of about $2.5 \times 10^{14} m^2$ with the half elimination time of about one month, the rate of deposition of beta activity per unit area per day may be estimated to be about 4.4×10^3 dpm/m^2 - day at 1 week, about 4.6×10^2 dpm/m^2 - day at 1 month and 25 dpm/m^2 - day at 3 months.

However the actual amount of precipitation may depend to a large extent on the type of nuclear detonation and the various meteorological conditions and the amount of rainfall. The composition of various radioactive nuclides seems to be different depending on the type of nuclear detonation and the condition at the time of detonation. Besides, some process of sorting or fractionation of nuclides seems to occur during transportation of activity from the site of detonation to the distant site of sampling. Oftentimes we have observed different decay rate of the gross activity of the samples collected in the suburb and in the urban industrial area, possibly because of the different chemical composition of the rainwater.

Of the various fission products produced at the time of nuclear detonation, the estimated yield of some of the radioactive nuclides which may be considered important from the health physics point of view are given in Table 1.

(Table 1)

If we assume 1.8×10^6 curies of Sr^{90} and 3.5×10^6 curies of Cs^{137} corresponding to 20 megaton TNT equivalent fast neutron fission of about 1,000 Kg of U^{238} were uniformly distributed over the earth's surface of about 5×10^8 km^2, the specific activity of Sr^{90} and Cs^{137} per unit area may be estimated at about 3.6 mc/km^2 or 3.6 mμc/m^2 of Sr^{90} and 7.0 mc/km^2 or 7.0 mμc/m^2 of Cs^{137}.

6. Np^{239} and U^{237}

Besides the fission products, several different radioactive nuclides may be produced by the capture of neutrons at the time of nuclear detonation.[17] Of these nuclides, neptunium-239 and uranium-237 which may be produced at the time of the fast neutron fission of uranium-238 may contribute considerably to the gross beta activity during the period up to about one and a half month after the nuclear detonation.

The neptunium-239 may be produced by the following reaction.

$$_{92}U^{238} + {}_0n^1 \longrightarrow {}_{92}U^{239} \xrightarrow[23.5m]{\beta^-} {}_{93}Np^{239} \xrightarrow[2.33d]{\beta^-} {}_{94}Pu^{239} \xrightarrow[2.4\times10^4y]{\alpha} \quad (50)$$

Although uranium-239 may be produced first by the intense neutron bombardment of uranium-238, it may be expected to decay out almost completely before reaching Japan.

Assuming that one atom of Np^{239} may produced per fission, the total activity of Np^{239} corresponding to the fast neutron fission of 1,000 Kg of uranium-238 may be estimated at about 2.4×10^{11} curies. Since neptunium-239 decays, exponentially while the gross activity of fission products may decrease according to $t^{-1.2}$ law, the ratio of the beta activity of Np^{239} to that of fission products may be given by

$$y_{(t)} = k\, e^{-\lambda t}\, .t^{1.2} \quad (51)$$

Table 1 ; Fission yield per 100 fissions

	Half-life	U^{235}		U^{238}	Pu^{239}	
		thermal	14 MeV	fast	thermal	fast
$_{38}Sr^{89}$	50.5 d	4.79	4.5	2.9	1.9	
$_{38}Sr^{90}$	28 y	5.77	4.5	3.2	2.3	2.2
$_{53}I^{131}$	8.06 d	3.1	4.3	3.2	3.8	
$_{55}Cs^{137}$	27 y, 28 y 30 y	6.15	5.7	6.2	5.24	6.6
$_{58}Ce^{144}$	282 d	6.0	3.3	4.9	5.29	
Cs^{137}/Sr^{90}		1.07	1.26	1.93	2.3	3.0

where K is a constant and t in unit of days.

Differentiating the above equation with respect to t, we may obtain

$$\frac{dy}{dt} = Kt^{0.2} e^{-\lambda t} (1.2 - \lambda t) \qquad (52)$$

where t is the time in days after fission, $\lambda = \frac{0.693}{2.33}$ /day and $K = \frac{2.4 \times 10^{11}}{2.6 \times 10^{11}}$. Since $\frac{dy}{dt} = 0$ at maximum $y_{(t)}$, the time to give a maximum in the above ratio of the beta activity may be given by

$$t = \frac{1.2}{\lambda} \qquad (53)$$

From the above equation, the activity of Np^{239} relative to that of the fission products may be estimated to reach a maximum at about 4 days after detonation, amounting to as much as about 60% of the total beta activity. The total activity of Pu^{239} corresponding to 2.4×10^{11} curies of Np^{239} may be estimated at about 6.4×10^{4} curies. If we assume this amount of Pu^{239} were uniformly distributed over the earth's surface of about 5×10^{14} m^2, the specific activity of Pu^{239} per unit area may be estimated to be about 128 $\mu\mu c/m^2$ or 128 $\mu c/km^2$.

The uranium-237 may be formed according to the following reaction.

$$_{92}U^{239} + {}_{0}n^{1} \rightarrow 2\,{}_{0}n^{1} + {}_{92}U^{237} \qquad (54)$$

Judging from the relative percentage of the activity U^{237} detected in the Bikini ash, it may be estimated that roughly about one atom of U^{237} might be formed per ten fission of uranium-238. Based upon this assumption, the total activity of uranium-237 per fast neutron fission of 1,000 Kg uranium-238 may be estimated at about 1.7×10^{10} curies. Substituting $\lambda = \frac{0.693}{6.7}$

into equation (52), the activity of U^{237} relative to that of the fission products may be estimated to reach a maximum at about 11.6 days after detonation, amounting to as much as about 50% of the total activity.

If we assume Np^{239} and U^{237} would contribute appreciably to the gross beta activity, the total beta activity as a function of the time t in days after detonation may be expressed by the following equation.

$$A_\beta = 2.6 \times 10^{11}\, t^{-1.2} + 2.4 \times 10^{11}\, e^{-\frac{0.693}{2.33} t} + 1.7 \times 10^{10}\, e^{-\frac{0.693}{6.7} t} \qquad (55)$$

The total beta activity represented by the above equation is shown in Fig.6. As can be seen in the figure, the total beta activity may be approximated in this case by the following equation during the period from a few days to about 30 days after the nuclear detonation.

$$A = A_o t^{-1.6}$$

where A_o is a constant and t the time after detonation.

(Fig 6)

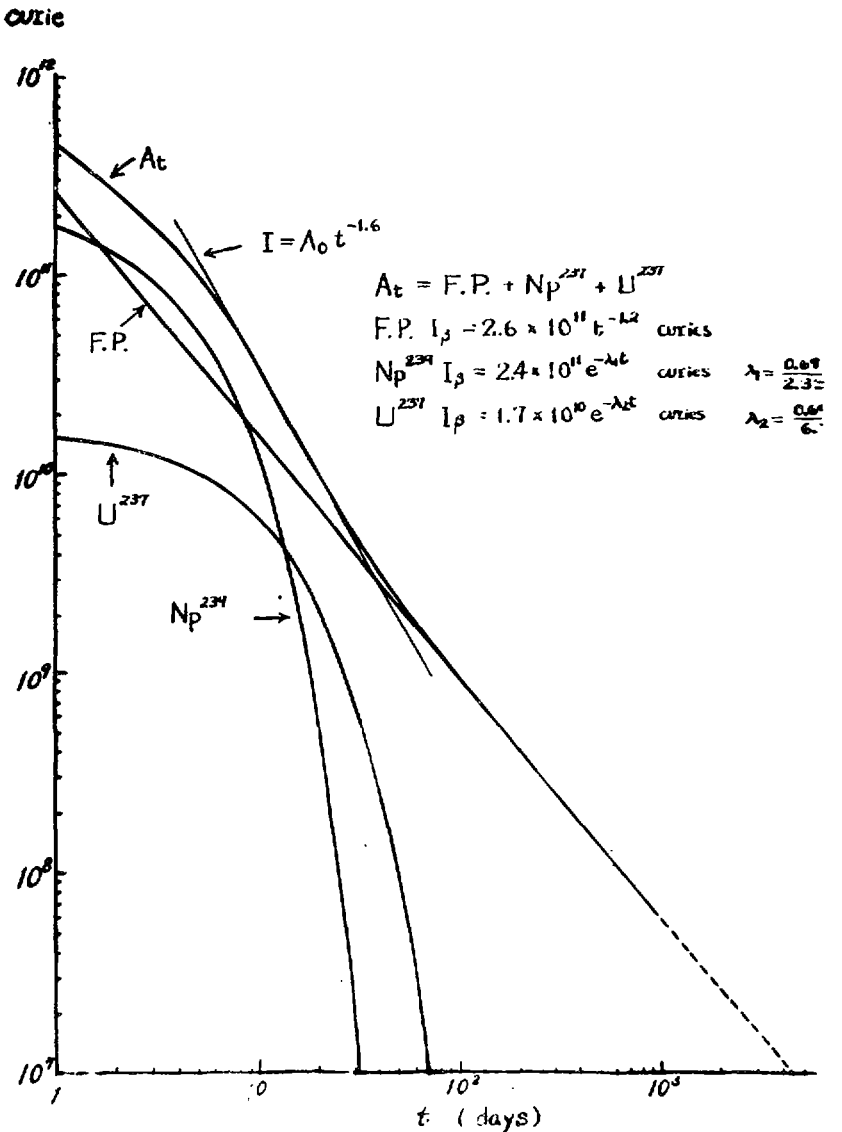

curie
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
At
$I = A_0 t^{-1.6}$
F.P.
U^{237}
Np^{239}
$A_t = F.P. + Np^{239} + U^{237}$
F.P. $I_\beta = 2.6 \times 10^{11} t^{-1.2}$ curies
Np^{239} $I_\beta = 2.4 \times 10^{11} e^{-\lambda_1 t}$ curies $\lambda_1 = \frac{0.69}{2.3}$
U^{237} $I_\beta = 1.7 \times 10^{10} e^{-\lambda_2 t}$ curies $\lambda_2 = \frac{0.69}{6.}$
1
10
10^2
10^3
t (days)

Fig. 6

III Discussion and Summary

In this paper some of the preliminary findings on the radioactive fallout due to nuclear testings during the period from March 1954 to the first half of 1955 are introduced. In order to check whether such a high radioactive fallout as observed during the period following a series of nuclear testings in Bikini in 1954, an order of magnitude estimation was made assuming the nuclear detonation to be of fission-fusion-fission type. Although in the above estimation a uniform distribution of tropospheric fallout over the entire surface of the northern hemisphere is assumed, the tropospheric fallout may occur in a belt-like pattern around the world starting from the site of nuclear detonation. Therefore, a strong radioactive fallout higher by one order of magnitude or more than the above estimates may be expected in some cases depending on the various meteorological conditions. In the case of stratospheric fallout, a difference in the radioactive fallout by a factor of about three may be expected depending on the position of jet stream above Japan. Usually a higher rate of radioactive fallout is observed at the central part of Japan when the jet stream is above the southern part of Japan. The fallout rate is estimated to be higher on the average by a factor of about three in the belt-like region around the earth between about N.30° and N.50° in the northern hemisphere. In Japan, at the Japan Sea side of the Japan mainland where more rainfall is usually observed the annual fallout rate is estimated to be two to three times higher than at the Pacific side.

Judging from these and other various meteorological factors, it would not be surprising to expect a radioactive fallout 10 - 100 times stronger than the estimates based upon the above order of magnitude estimation. The degree of radioactive fallout may also be cousiderably different locally depending upon the local

meteorological conditions and the amount of rainfall. In some extreme cases we have observed during the initial period of preliminary observation a large difference of radioactivity in the rainwater collected at two different side of the building. When a strong wind is blowing from one direction the initial highly radioactive rainfall is collected fully on one side of the building facing the wind, while not even a drop on the other side.

Following the unusally highly radioactive rainfall in the middle of May 1954 in Osaka - Kyoto district we have caught a frog in the rice field in the suthern suburb of Osaka and examined for the radioactive contamination of the internal organs. In Fig.7, the results of the examination are shown in unit of cpm/g which is the gross beta activity detected excluding the potassium activity. ←(Fig.7)

Since a considerable number of people on the rainwater in the isolated small island and in the mountain district, in order to see what level of grossactivity would be permissible for drinking water the author tried to estimate the total activity of major nuclides which may be considered most important from the health physics point of view and the fraction of those activities of individual nuclides to the gross beta activity of fission products assuming the fission yield of the fast neutron fission of U^{238} as given in Table 1.

The results of these estimations are given in Table 2 and 3. The total activities in these tables were computed for the 20 megaton equivalent fission of U^{238}. In Table 4 the maximum permissible daily intakes of major nuclides are estimated based upon the occupational maximum pernissible concentration of drinking water given in ICRP (1958)[18] assuming the amount of water taken per day to be about 2.2 litres on the average.

(Table. 2)
(Table. 3)
(Table. 4)

frog (May 25, 1954)

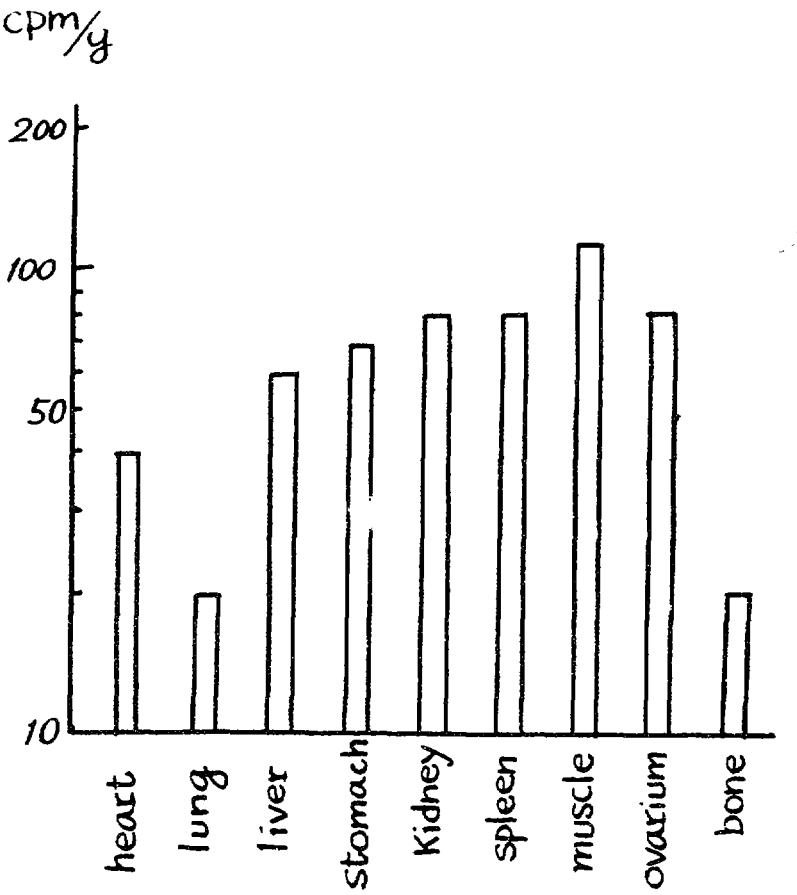


Fig. 7

Table 2 ; Total activity corresponding to 20 megaton equivalent fission of U^{238}

Nuclide	Activity	1w	1m	3m	1yr
Sr^{89}	3.3×10^8c (1.0)	3×10^8c (0.9)	2.2×10^8c (0.66)	9.6×10^7c (0.29)	2.2×10^6c (6.6×10^{-3})
Sr^{90}	1.8×10^6c (1.0)	1.8×10^6c ($\sim$ 1.0)	1.8×10^6c ($\sim$ 1.0)	1.8×10^6c (0.99)	1.76×10^6c (0.976)
I^{131}	2.3×10^9c (1.0)	1.3×10^9c (0.55)	1.8×10^8c (7.7×10^{-2})	1.1×10^6c (4.5×10^{-4})	6×10^{-5}c (2.5×10^{-14})
Cs^{137}	3.5×10^6c (1.0)	3.5×10^6c ($\sim$ 1.0)	3.5×10^6c ($\sim$ 1.0)	3.5×10^6c (0.99)	(3.41×10^6c) (0.976)
Ce^{144}	9.8×10^7c (1.0)	9.8×10^7c ($\sim$ 1.0)	9.1×10^7c (0.93)	7.8×10^7c (0.8)	4×10^7c (0.41)

Table 3; Fraction to gross beta activity of fission productions mixture

Nuclide	3 d	1 w	1 m	3 m	1yr
F.P.β	6.9×10^{10}c	2.6×10^{10}c	4.5×10^{9}c	1.2×10^{9}c	2.2×10^{8}c
Sr^{89}	4.8×10^{-3}	1.2×10^{-2}	*4.9×10^{-2}	*8×10^{-2}	*1.0×10^{-2}
Sr^{90}	2.6×10^{-5}	6.9×10^{-5}	4×10^{-4}	1.5×10^{-3}	*8×10^{-3}
I^{131}	*3.3×10^{-2}	*5×10^{-2}	*4×10^{-2}	9.2×10^{-4}	2.7×10^{-13}
Cs^{137}	5.1×10^{-5}	1.35×10^{-4}	7.8×10^{-4}	2.9×10^{-3}	*1.55×10^{-2}
Ce^{144}	1.4×10^{-3}	3.8×10^{-3}	2×10^{-2}	*6.5×10^{-2}	*1.8×10^{-1}

Table 4 ; ICRP (1958)

Nuclide	Effective Half-life	Critical Organ	Maximum Permissible Burden (μc)	$(M.P.C)_w$ (μc/cc)	Daily Intake (μc)
Sr^{89}	50.4 d	bone	4	10^{-4}	2.2×10^{-1}
Sr^{90}	6.4×10^{3} d	bone	2	10^{-6}	2.2×10^{-3}
I^{131}	7.6 d	thyroid	0.7	2×10^{-5}	4.4×10^{-2}
Cs^{137}	70 d	whole body	30	2×10^{-4}	4.4×10^{-1}
Ce^{144}		G.I.		10^{-4}	2.2×10^{-1}
	243 d	bone	5	8×10^{-2}	1.76×10^{2}
Np^{239}		G.I.		10^{-3}	2.2
	2.33 d	bone	30	30	6.6×10^{4}

As can be seen in these tables, the most important nuclide from the health physics point of view in the radioactive contamination at about one week after the nuclear detonation may be assumed to be the radioactive iodine I^{131} which may be estimated to exist in a few percent of the gross beta activity of fission products during the period from a few days to about one month after the nuclear fission of U^{238}.

During the initial period following the nuclear detonation the relative percentage of such induced activities as Np^{239} or U^{237} may be higher than that of the radioactive iodine, but because of the lower maximum permissible concentration of the radioactive iodine which concentrate in thyroid I^{131} may still be considered the most critical nuclide. At about three months Sr^{90} and Ce^{144} may be considered critical. With the lapse of time the gross activity of the fission products mixture may decrease greatly and at one year the contribution of I^{131} to the gross activity may be considered negligible, but the relative importance of such long lived fission product as Sr^{90} and Cs^{137} may gradually increase. Assuming I^{131} to be the only critical nuclide at about one week, the gross beta activity of fission products mixture corresponding to the occupational maximum permissible concentration of I^{131} may be estimated to be about 400 μμc/cc and even if we assume the contribution of such induced activities as Np^{239} and U^{237} with relatively short half lives to be about 60% of the total activity it may be estimated at about 1,000 μμc/cc.

During the initial period following the nuclear detonation, the relative importance of the individual nuclides other than I^{131} may be lower than I^{131}, but the combined effects of all other nuclides may not be negligible. Taking such factors into consideration, it may be safer to assume about one hundredth of the above value or 4 - 10 μμc/cc as the permissible concentration of gross activity of the mixture of beta and gamma emitters

we must pay attention to such alpha emitters as Pu^{239} which may be included usually in a very small percentage. Although the permissible concentration for such a mixture of various radioactive nuclides should be decided after the radiochemical analysis, it is oftentimes impossible to identify relative percentage of all the nuclides as rapid and as accurate as required to caution the people about the consumption of contaminated water and food.

In such cases, because of the various unknown factors included in the estimation, the author feels that the permissible level for the general people should not be higher than 1μμc/cc as measured with the gross activity during the period from a few days to about one week following the nuclear detonation when we usually observe strongly radioactive rainfall in Osaka Japan. After the strongly radioactive rainfall, the radioactive concentration in the air is usually observed to decrease considerably.

However, assuming the intake of the radioactivity by inhalation to be about one tenth of that by ingestion and the daily intake of water to be about 2.2 litres, the level of gross activity of 1 μμc/cc given in the above may correspond to about 2,000 μμc of daily intake of grossactivity by ingestion. In case of repeated nuclear testings with relatively short intervals the relative percentage of such critical nuclide as I^{131} may be increased. Taking such possibilities into consideration, it would be better to set as the first warning level during the initial period the maximum permissible daily intake of grossactivity by ingestion through water and food for general people at about 1,000 μμc/day or lower depending on the relative contribution of critical nuclides. If the nuclear testings should be continued indefinitely for the future, the relative contribution of such long lived and critical nuclides as Sr^{90}, Cs^{137} and Pu^{239} from the slow stratospheric fallout may be expected to increase gradually. Therefore it seems to be extremely important to continue the analysis of such long

lived nuclides in the rainwater for the future especially from the health physics point of view.

The above level of total intake of gross activity 1,000 μμc per day may be considered to correspond to roughly about 100 cpm/l for the people using the rainwater as the only source of drinking water, assuming the major route of intake of activity to be only the drinking water of 2.2 litres per day and the efficiency of counting the activity to be about 10%.

However, in case the extremely highly radioactive rain should fall all over the entire country, there may be the possibility that the above level might be exceeded everywhere simultaneously. Preparing for such an emergency situation it would be advisable to set several different stages of warning levels of radioactive contamination for the people and to try to have the general public understand the meaning and significance of those levels during the normal time prior to the arrival of such an emergency.

From what is discussed in the above, even at this day it may be said definitely for the general public that the rainwater should not be used as the only source of drinking water continuously without filtering, and that it is advisable to try to avoid the initial highly radioactive part of rainfall in collecting the rainwater for drinking and cooking purposes, if possible.

The radioactive contamination on the surface of the vegetables by direct exposure to radioactive rain was observed to be decontaminated by about 90% or more by washing thoroughly with 0.1 - 1N dilute hydrochloric acid or with the acetic acid or citric acid and the surface active agents such as soapless soap and the chelating agents such as EDTA-Na, DETPA-Na or nitrilo-triacetate-Na. With a mixed filter of bone charcoal and clay 90 - 99% of the fission products activity was observed to be eliminated from the contaminated water.

Acknowledgement

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Studies On the Radioactive Contamination due to Nuclear Detonations III

(On the Method of Estimating the Probable time of Nuclear Detonation from the Measurements of Gross-activity)

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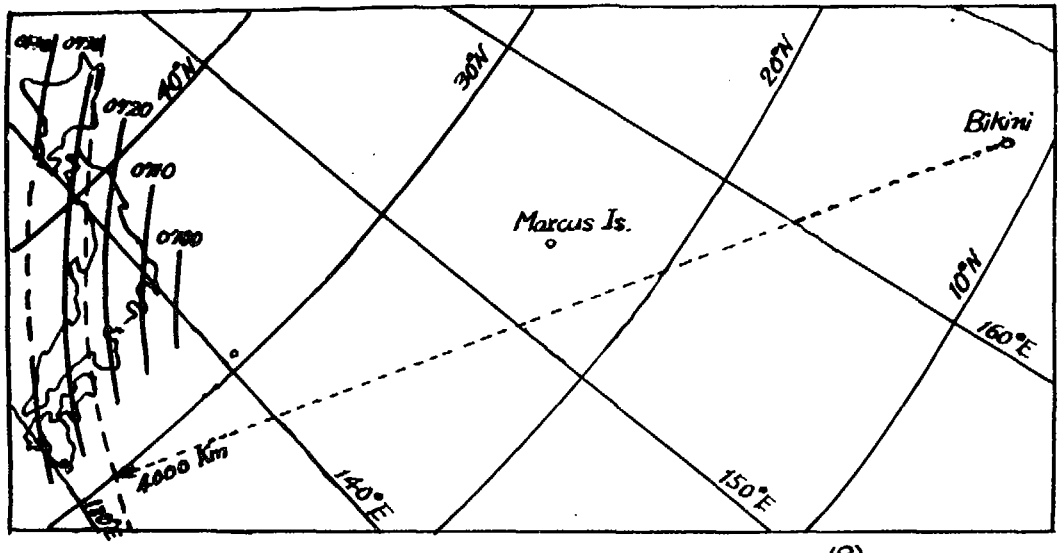
On the Method of Estimating the Probable Time of Nuclear Detonation from the Measurements of Gross-activity

Since it has been observed in Spring of 1954 that a considerable amount of fission products mixture fell with the rain following a large scale nuclear detonation conducted in Bikini area in the South Pacific by the United States Atomic Energy Comission, it has become important, especially from the health physics standpoint, to estimate the effective average age of the fission products mixture after the nuclear detonation. If the energy transferred to the atmospheric air at the time of nuclear detonation is large enough (order of 1 megaton at the distance of about 4000km), the probable time and test site of nuclear detonation may be estimated with considerable accuracy, as shown in Fig.1 and in Table 1, from the records of the pressure wave caused by the detonation in the microbarographs at different meteorological stations.[1][2] Even in this case, in order to estimate the possible corelation between the artificial radioactivity observed in the rain and the probable detonation, it is oftentimes desirable to estimate the effective ago of the fission products mixture in the rain from the decay measurement of the radioactivity.

(Fig.1)

(Table 1)

Especially if the energy transferred to the atmospheric air by the detonation is small and the test site is too remote, the identification of the pressure wave caused by the detonation with sufficient accuracy would be rather difficult, judging from the sensitivity of the past method of detection. Even in such cases, it was occasionally observed that a considerable amount of radioactivity fell with the rain at remote places depending on the meteorological conditions. Therefore, it was considered rather important to estimate the probable date of nuclear



(2)

Fig. 1 : Isochronous chart on 1 March 1954

Table 1. Observed results of pressure waves
(After Japan Meteorological Agency, Tokyo)

No.	Year	Probable Time				Amplitude (mb)	Period (min)	Duration (min)	Test site
				h	m				
1	1954	II	28	22	07	0.3	5	30	Bikini Atoll
2	"	III	26	21	22	0.2	4	30	"
3	"	IV	25	21	28	0.2	4	30	"
4	"	V	4	21	29	0.2	4	30	"
5	1955	XI	22	8	08	0.1	?	10	Near Mongol
6	1956	V	20	21	35	0.2	3-4	40	Eniwetok
7	"	V	27	21	13	0.2	2	40	"
8	"	VI	25	21	24	0.2	2	50	"
9	"	VII	2	21	16	0.1	1	30	"
10	"	VII	8	21	13	0.3	2	40	"
11	"	VII	10	21	10	0.6	5	40	"
12	"	VII	20	20	59	0.5	3	55	"
13	"	VII	21	21	24	0.1	1	20	"
14	"	VIII	30	10	45	0.04	2	45	SW Siberia
15	"	X	17	12	45	0.2	1	30	"
16	1957	IV	3	13	-	0.02	?	10	"
17	"	IV	10	11	59	0.2	2	20	"
18	"	IV	16	11	01	0.1	2	30	"
19	"	IX	24	14	26	0.1	1-4	30	North polar region
20	"	X	6	14	29	0.3	3	30	"

No.	Year	Probable Time			Amplitude (mb)	Period (min)	Duration (min)	Test site
			h	m				
21	1958	II 23			0.3	1	40	North polar region
22	"	II 27	13	26	0.5	1-2	35	"
23	"	II 27	15	57	0.2	1	20	"
24	"	III 14	13	30	0.1	?	10	"
25	"	III 14	14	07	0.1	?	10	"
26	"	III 21	14	25	0.2	1	30	"
27	"	V 12	21	35	0.1	1-3	30	Eniwetok
28	"	V 26	5	08	0.1	1	20	"
29	"	VI 10	20	46	0.1	1	20	Bikini Atoll
30	"	VI 14	20	47	0.1	1	40	Eniwetok
31	"	VI 14	21	35	0.2	1-3	35	"
32	"	VI 27	20	51	0.2	2	20	"
33	"	VI 27	21	36	0.4	2	15	"
34	"	VI 28	22	29	0.9	2-5	40	"
35	"	VII 2	20	51	0.1	1	15	Bikini Atoll
36	"	VII 5	21	39	0.1	1	20	Eniwetok
37	"	VII 12	6	41	0.5	2-4	40	"
38	"	VII 26	23	34	0.2	1-3	40	"
39	"	VIII 12	15	11	0.1	4	35	North polar region
40	"	IX 30	13	16	0.2	2	20	"
41	"	IX 30	15	19	0.2	3	25	"
42	"	X 12	13	14	0.1	3	35	"
43	"	X 15	13	18	0.2	3	40	"
44	"	X 19	15	13	0.3	3	30	"
45	"	X 20	11	49	0.1	1	15	"
46	"	X 22	13	43	0.2	2	35	"
47	"	X 24	13	24	0.1	3	30	"
48	"	X 25	13	05	0.1	1	15	"

detonation from the measurement of the artificial radioactivity of the rain, in order to assess the relative hazards of the gross activity of the rain from the health physics point of view.

In this paper, various methods of estimation of the probable time of nuclear detonation from the gross activity of the rain are introduced and discussed based upon the past seven years experiences of the observation of the artificial radioactivity of the rain by the author and his associates in Osaka.

(I) Method of Dating a nuclear detonation from the measurement of the decay of gross-activity in the rain or dust.

According to Way and Wigner[3] the composite radioactive decay resulting from nuclear fission may be expressed by the following type of power function of the time τ after the nuclear fisson.

$$I = A_0 \tau^{-\alpha} \qquad (1)$$

where I is the activity at time τ days after the nuclear fission, Ao a theoretical constant corresponding to the activity at unit time after fission, α another constant whose theoretical value has been given to be 1.2 for the mixture of fission products. Substituting the relation $\tau = t - t_0$ into equation (1), we may obtain

$$I = A_0 (t - t_0)^{-\alpha} \qquad (2)$$

where t is the time of measurement of gross activity, t_0 the time or date of the nuclear fission. According to our past experiences somewhat different values of α were obtained even with the same sample depending on the degree of absorption of radiation by a layer between the source and the counter or by the counter window itself. Although the value of α was observed to vary from 0.8 to 1.9 depending on various factors such as the method of detection, possible sorting or fractionation of nuclides at the time of sampling or during natural transportation from the detonation site to the place of sampling or the type of nuclear

detonation, meteorological conditions and others, the above relation was observed to hold approximately with our method of detection for the period from a few days to about 30 - 50 days after nuclear detonation with a constant value of α even in case when such nuclides as U^{237} or Np^{239} are included besides the ordinary fission products in the radioactive dust.

From about 50 - 60 days to about 200 - 300 days after the nuclear detonation the above relation may still be assumed to hold approximately, but with somewhat different constant value of α which is oftentimes observed to be smaller than the initial value of α in case the initial value of α is larger than 1.2. This may probably due to the decaying out of such nuclides with relatively short half-lives as uranium-237, neptinium-239 and other transuranium elements or some other induced radioactivity of relatively short half-lives which might have been produced at the time of nuclear detonation but may not be considered so called ordinary fission products. When an appreciable amount of U^{237} or Np^{239} was detected in the rain besides the ordinary fission products, the apparent value of α during the initial period of a few days to about 30 days was usually observed to be larger than 1.2.

In view of these findings, various methods of dating the nuclear detonations described in the below have been derived, assuming, a priori, that the Way-Wigner relation holds for the composite radioactivity resulting from nuclear detonations or even from what is usually called H-bomb tests.
The value of α may be expected to vary depending on the type of nuclear detonation and other factors as mentioned above.

Method (I)

Assuming the Way-Wigner relation as expressed by the equation (1) or (2), we may obtain the following relations

$$I_1 = A_o (t_1 - t_o)^{-\alpha} \qquad (3)$$

$$I_2 = A_o (t_2 - t_o)^{-\alpha} \qquad (4)$$

$$I_3 = A_o (t_3 - t_o)^{-\alpha} \qquad (5)$$

where I_1, I_2 and I_3 are the activeties measured at the times t_1, t_2 and t_3, and t_o is the time of nuclear detonation.

If we could obtain t_1, t_2 and t_3 corresponding to the following relation by either the direct measurement or by the extrapolation or interpolation of the decay curve of the gross-activity,

$$\frac{I_1}{I_2} = \frac{I_2}{I_3} = R \qquad (6)$$

the following relation holds,

$$I_2^2 = I_1 I_3 \qquad (7)$$

where R can be any constant arbitrarily chosen.
Substituting the relations (3) - (5) into (7), we may obtain

$$(t_2 - t_o)^2 = (t_1 - t_o)(t_3 - t_o) \qquad (8)$$

Solving the above equation with respect to t_o, we obtain the probable time of detonation t_o as a function of t_1, t_2, and t_3.

$$t_o = \frac{t_1 t_3 - t_2^2}{t_1 + t_3 - 2t_2} \qquad (9)$$

When t_o is estimated, the corresponding value of α may be easily obtained from the relations (3) - (5). However, considering the relation (6), α may be expressed by the following

equation as a function of t_1, t_2, t_3 and R.

$$\alpha = \ln R \,/\, \ln \left(\frac{t_3 - t_2}{t_2 - t_1} \right) \qquad (10)$$

With this method, somewhat different values of t_o and α may be obtained from a different set of triplet t's corresponding to the relation (6).

Therefore, it seems to be safer to estimate the mean values of t_o and α by treating statistically different values of t_o and α obtained from the different sets of t's extrapolated ~~or interrelated~~ or interpolated corresponding to the relation (6) from a series of measurements of gross-activity for a proper time interval.

In case the most probable estimate of t_o is first obtained from the above method, or if t_o is known previously, the following analytical method may be used to estimate the most probable values of α and A_o.

Assuming t_o is known, for the sets of measurements (I_1, t_1), (I_2, t_2),------, (I_i, t_i),------, (I_n, t_n), since we may obtain from the Way-Wigner relation the following relation,

$$\ln I_i = \ln A_o - \alpha \ln (t_i - t_o) \qquad (11)$$

the experimental error may be expressed by

$$S = \sum \left[\ln I_i - \ln A_o + \alpha \ln \tau_i \right]^2 \qquad (12)$$

where $\tau_i = t_i - t_o$ and $i = 1,2,\text{------}, n$.

Since S may be considered to involve the errors in estimating α and A_o, the following condition may be satisfied for minimum error in α and A_o.

$$\frac{\partial S}{\partial \alpha} = 0, \qquad \frac{\partial S}{\partial A_o} = 0 \qquad (13)$$

Solving the above equations with respect to α and A_o, we may obtain,

$$\alpha = \frac{\sum(\ln I_i)\sum(\ln \iota_i) - n\sum(\ln I_i)(\ln \iota_i)}{n\sum(\ln \iota_i)^2 - \left[\sum \ln \iota_i\right]^2} \qquad (14)$$

$$A_o = \exp\left[\frac{\sum(\ln I_i)\sum(\ln \iota_i)^2 - \sum(\ln I_i)(\ln \iota_i)\sum(\ln \iota_i)}{n\sum(\ln \iota_i)^2 - \left[\sum \ln \iota_i\right]^2}\right] \qquad (15)$$

where $\iota_i = t_i - t_o$.

In case the most probable estimate of α is first known, since we may obtain from the Way-Wigner relation the following expression

$$I_i^{-\frac{1}{\alpha}} = A_o^{-\frac{1}{\alpha}}(t_i - t_o) \qquad (16)$$

the experimental error may be expressed by

$$S = \sum\left[y_i - A_o^{-\frac{1}{\alpha}}(t_i - t_o)\right]^2 \qquad (17)$$

where $y_i = I_i^{-\frac{1}{\alpha}}$ corresponding to t_i and $i = 1, 2, \text{———}, n$. For minimum error in t_o and A_o, the following condition may be satisfied.

$$\frac{\partial S}{\partial t_o} = 0 \qquad \frac{\partial S}{\partial A_o} = 0 \qquad (18)$$

Solving the above equations with respect to t_o and A_o, we may obtain,

$$t_o = \frac{\sum y_i \sum(t_i)^2 - \sum y_i\, t_i \sum t_i}{\sum y_i \sum t_i - n\sum y_i\, t_i} \qquad (19)$$

$$A_o = \left[\frac{(\ t_i)^2 - n\ (t_i)^2}{y_i\ \ t_i - n\ \bar{y}_i t_i}\right]^{\alpha} \qquad (20)$$

where $y_i = I_i^{-\frac{1}{\alpha}}$ and I_i is the activity at time t_i.

Method (II)

This method may be considered a modified method of Method (I). Assumong the Way-Wigner relation, we may obtain the following relation.

$$I = A_o (t - t_o)^{-\alpha} \tag{21}$$

$$I - \Delta I = A_o (t + \Delta t - t_o)^{-\alpha} \tag{22}$$

Dividing the equation (22) by the equation (21), we may eliminate the constant A_o and obtain

$$\left(\frac{I}{I - \Delta I}\right)^{\frac{1}{\alpha}} = \frac{t + \Delta t - t_o}{t - t_o} \tag{23}$$

Subtracting unity from both sides of the above equation,

$$\left(\frac{I}{I - \Delta I}\right)^{\frac{1}{\alpha}} - 1 = \frac{\Delta t}{t - t_o} \tag{24}$$

If we obtain the value of Δt corresponding to a constant value of ΔI or a constant ratio of the activity R by extrapolation or interpolation from a series of measurements, the left side of the above equation will be a constant K, i. e.

$$\left(\frac{I}{I - \Delta I}\right)^{\frac{1}{\alpha}} - 1 = K \tag{25}$$

where

$$\frac{I}{I - \Delta I} = R \tag{26}$$

is a constant arbitrarily chosen as in the relation (6) in the Method (I). Substituting the constant K of equation (25) into equation (24), and rearranging, we may obtain

$$\Delta t = K (t - t_o) \qquad (27)$$

Since the equation (27) involves only two unknown constants t_o and K, if we obtain two or more sets of corresponding values of t and Δt, we may easily estimate the value of t_o and K or α as described in Method (I).

Yamazaki and Kaneko[4] used a graphical method in estimating the value of t_o and K or α. Assuming t_o and K are constants, in equation (27), the relation between t and Δt will be linear, therefore by plotting Δt's against t estimated from a smothed out decay curve corresponding to the condition (26), a straigth line may be obtained. By extrapolating this straight line to a point corresponding to $\Delta t = 0$ we may obtain the value of t_o and from the slope of the line the value of K may be estimated. When the value of K is estimated, the corresponding value of α may be obtained from the relations (25) and (26) since the ratio of the activity R is an arbitrarily chosen known constant. Therefore, α may be given by

$$\alpha = \ln R / \ln (1 + K) \qquad (28)$$

However, with this graphical method the error in the estimation of t_o will be large because a slight difference in the slope of the straight line drawn by sight will give a considerably different value of t_o at the time of extrapolation and the author found it oftentimes rather difficult to estimate t_o and α more objectively without any previous knowledge about the probable time of nuclear detonation. Therefore, the author has been trying to estimate t_o and α more objectively with the following analytical method and used the graphical method as a subsidiary method to check if any serious mistakes were made in the calculation by comparing the graphical plot of Δt vs t and the straight line estimated by the analytical method.

Assuming the relation as expressed by the equation (27) holds for a series of estimated values $(\Delta t_1, t_1)$, $(\Delta t_2, t_2)$, ------, $(\Delta t_i, t_i)$, ------, $(\Delta t_n, t_n)$ corresponding to the condition (26), the error in the estimation may be expressed by

$$S = \sum \left[\Delta t_i - K(t_i - t_o) \right]^2 \qquad (29)$$

where

$$K = R^{\frac{1}{\alpha}} - 1 \qquad (30)$$

and

$$R = \frac{I_i}{I_i - \Delta I_i} = \text{constant} , \quad (i=1,2,\cdots\cdots, n) \qquad (31)$$

For minimum error in t_o and K, the following condition may be satisfied.

$$\frac{\partial S}{\partial t_o} = 0, \quad \frac{\partial S}{\partial K} = 0 \qquad (32)$$

Solving the above equations as in the Method (I), we may obtain,

$$t_o = \frac{\sum \Delta t_i \sum (t_i)^2 - \sum (\Delta t_i)(t_i) \sum t_i}{\sum \Delta t_i \sum t_i - n \sum (\Delta t_i)(t_i)} \qquad (33)$$

$$K = \frac{\sum \Delta t_i \sum t_i - n \sum (\Delta t_i)(t_i)}{(\sum t_i)^2 - n \sum (t_i)^2} \qquad (34)$$

Since $K = R^{\frac{1}{\alpha}} - 1$, α may be expressed as

$$\alpha = \ln R \,/\, \ln \left[1 + \frac{\sum \Delta t_i \sum t_i - n \sum (\Delta t_i)(t_i)}{(\sum t_i)^2 - n \sum (t_i)^2} \right] \qquad (35)$$

where $R = \frac{I_i}{I_i - \Delta I_i}$ is a constant, corresponding to which Δt_i should be estimated from the experimental decay curve.

Method (III)

With the foregoing methods, we have to estimate the time corre-

sponding to a reduction of gross-activity by a certain constant ratio as indicated by the condition (6) or (26). Since it is rather difficult to obtain the corresponding time by the direct measurement with the ordinary equipment, we may oftentimes have to estimate the time by extrapolation or interpolation from the smoothed out decay curve. However, since the time interval between the measurements can be arbitrarily chosen and the corresponding rate of decrease of radioactivity may be obtained from the direct measurement, the author tried to develop a method with which the values of t_o and α may be estimated from the rate of decrease of gross-activity corresponding to a certain constant ratio of time interval between the measurements of activity.

Assuming the Way-Wigner relation, we have the following relations for a set of triplet observed values (I, t), $(I_1, t + \Delta t)$, $(I_k, t + k\Delta t)$

$$I = A_o (t - t_o)^{-\alpha} \tag{36}$$

$$I_1 = I - \Delta I = A_o (t + \Delta t - t_o)^{-\alpha} \tag{37}$$

$$I_k = I - \Delta I_k = A_o (t + k\Delta t - t_o)^{-\alpha} \tag{38}$$

where I, I_1 and I_k are the activities corresponding to the time of measurements t, $t + \Delta t$, and $t + k\Delta t$, and t_o the time or date of nuclear detonation.

Dividing the equation (37) by the equation (36) and subtracting unity from both sides of the equation, we may obtain

$$\left(\frac{I}{I_1}\right)^{\frac{1}{\alpha}} - 1 = \frac{\Delta t}{t - t_o} \tag{39}$$

Similarly, dividing the equation (38) by the equation (36) and subtracting unity from both sides of the equation, we may obtain

$$\left(\frac{I}{I_k}\right)^{\frac{1}{c}} - 1 = \frac{k\Delta t}{t - t_o} \tag{40}$$

Dividing the equation (40) by the equation (39), we have

$$\frac{\left(\frac{I}{I_k}\right)^{\alpha} - 1}{\left(\frac{I}{I_1}\right)^{\alpha} - 1} = k \qquad (41)$$

Since $\frac{I}{I_1}$ and $\frac{I}{I_k}$ and k can be obtained from the direct measurements and k is an arbitrarily chosen constant prior to measurements, α may be estimated from the equation (41). When α is estimated, the corresponding value of t_o can be estimated either from equation (39) or from equation (40), since all other variables except t_o in these equations are known.

However, it is not always very easy to estimate the value of α from the equation (41). Therefore, the author attempted to obtain the approximate estimation of α from the chart as shown in Fig. 5 or Fig. 6 which was constructed according to the following equation corresponding to the equation (41),

$$\frac{Y^{\frac{1}{\alpha}} - 1}{X^{\frac{1}{\alpha}} - 1} = k \qquad (42)$$

where $X = \frac{I}{I_1}$ and $Y = \frac{I}{I_k}$.

The value of k can be any constant theoretically, but for the convenience of estimation the chart in Fig.2 was compauted for k = 2, and that in Fig. 3 for k = 3. Therefore if we failed to measure the activity at the time corresponding to k = 2 or k = 3, we may have to estimate first the corresponding activities from the decay curve in order to use the above charts, otherwise we may have to estimate α directly from the equation (41). The numerical values, based on which the charts in Fig.2 or 3 were constructed, are given in Tables 2 and 3. As can be seen

(Fig.2)
(Fig.3)

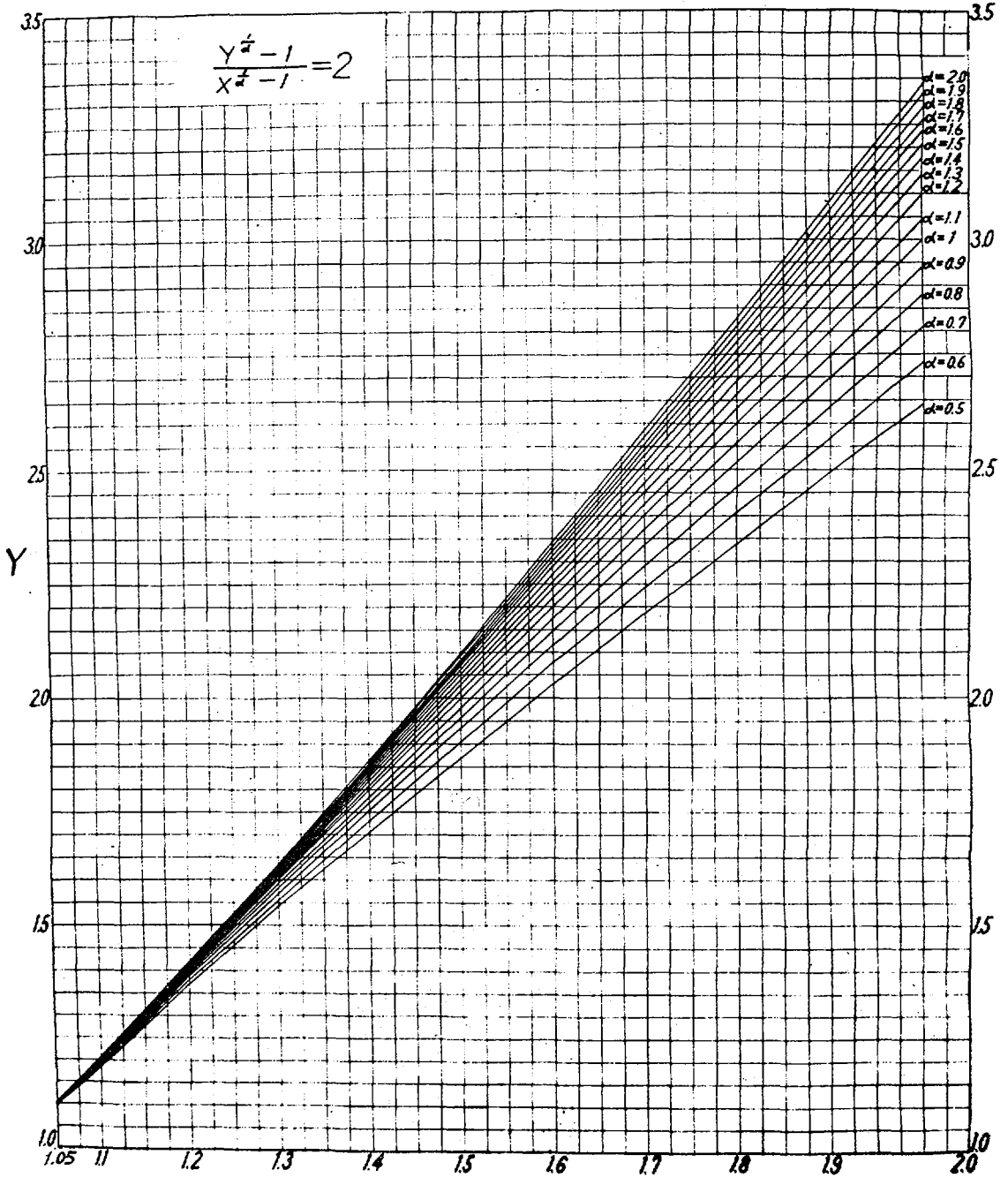

$\frac{Y^{\frac{1}{\alpha}}-1}{X^{\frac{1}{\alpha}}-1}=2$
α=2.0
α=1.9
α=1.8
α=1.7
α=1.6
α=1.5
α=1.4
α=1.3
α=1.2
α=1.1
α=1
α=0.9
α=0.8
α=0.7
α=0.6
α=0.5
Y
X
3.5
3.0
2.5
2.0
1.5
1.0
1.05
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2.0

Fig 2 -(a)

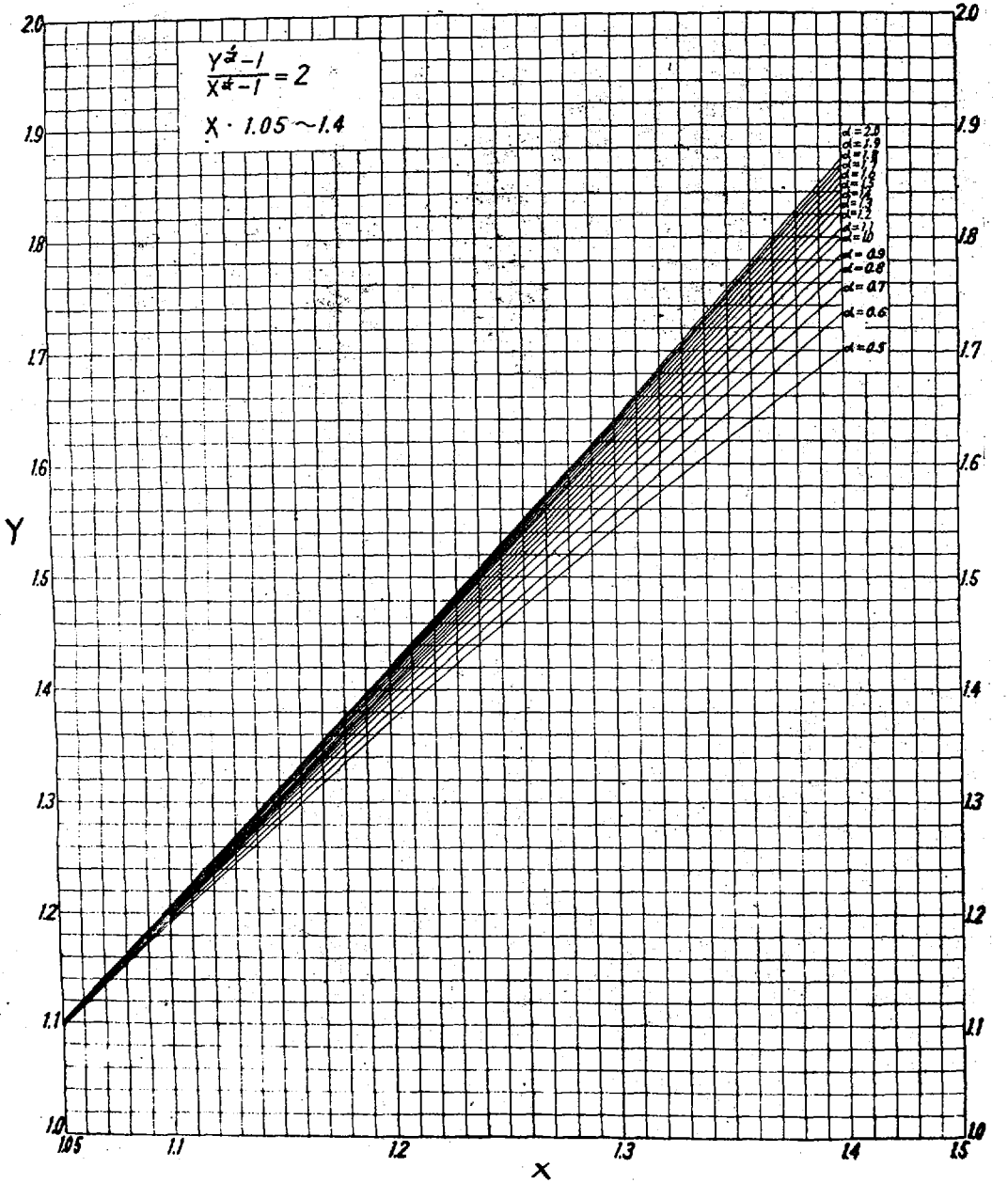

$\frac{Y^{\alpha}-1}{X^{\alpha}-1}=2$
X · 1.05 ~ 1.4
α = 2.0
α = 1.9
α = 1.8
α = 1.7
α = 1.6
α = 1.5
α = 1.4
α = 1.3
α = 1.2
α = 1.1
α = 1.0
α = 0.9
α = 0.8
α = 0.7
α = 0.6
α = 0.5
Y
X
2.0
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.0
1.05
1.1
1.2
1.3
1.4
1.5

Fig. 2 —(b)

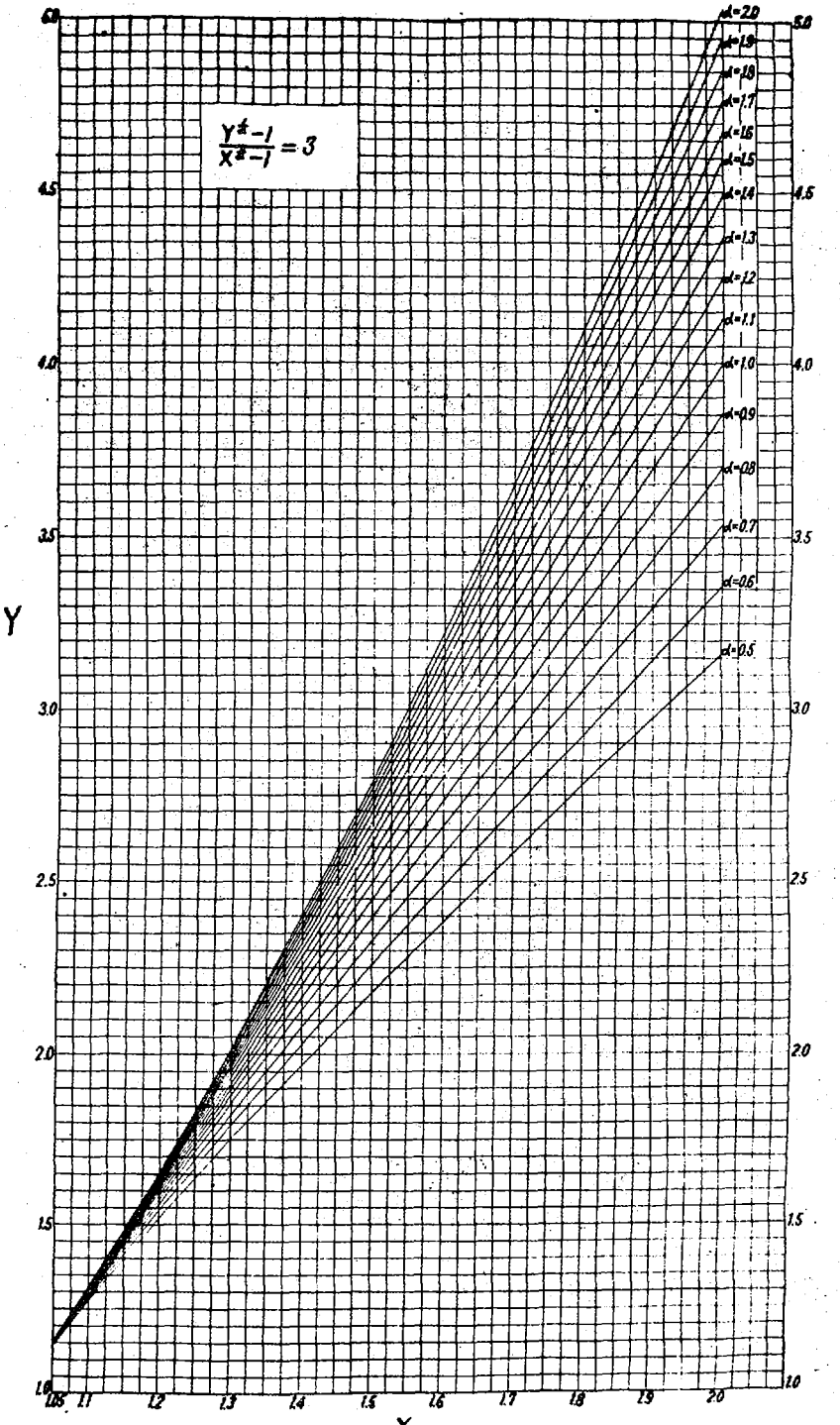

$\frac{Y^{\alpha}-1}{X^{\alpha}-1}=3$
α=2.0
α=1.9
α=1.8
α=1.7
α=1.6
α=1.5
α=1.4
α=1.3
α=1.2
α=1.1
α=1.0
α=0.9
α=0.8
α=0.7
α=0.6
α=0.5
Y
X
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
1.05
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2.0

Fig. 3 – (a)

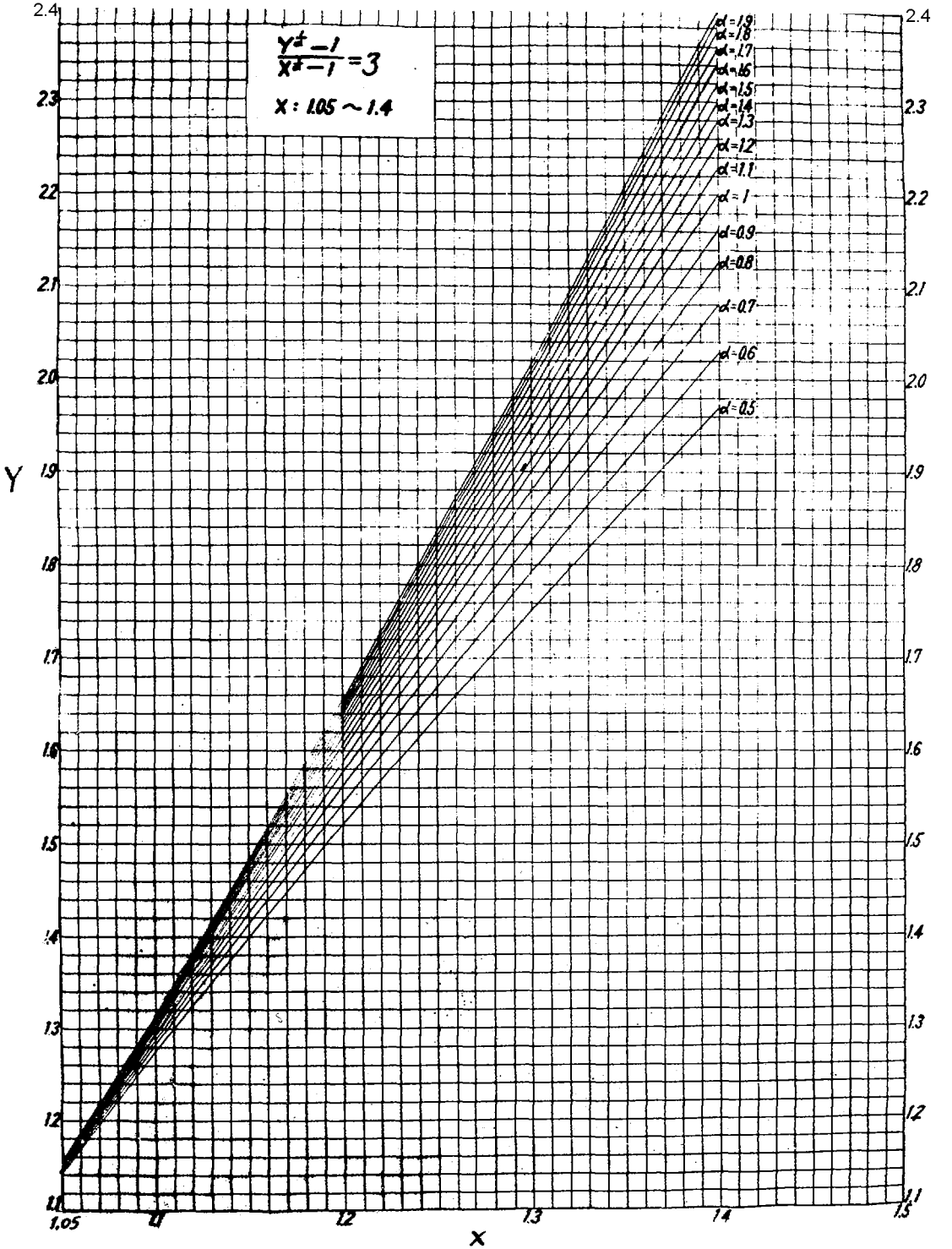

$\frac{Y^2-1}{X^2-1}=3$
X : 1.05 ~ 1.4
α=1.9
α=1.8
α=1.7
α=1.6
α=1.5
α=1.4
α=1.3
α=1.2
α=1.1
α=1
α=0.9
α=0.8
α=0.7
α=0.6
α=0.5
Y
X
2.4
2.3
2.2
2.1
2.0
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
1.05
1.1
1.2
1.3
1.4
1.5

Fig. 3 - (b)

Table 2 : $Y = \left[1 + 2\left(X^{\frac{1}{\alpha}} - 1\right)\right]^{\alpha}$

α \ X	1.05	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2.0
0.5	1.0977	1.1916	1.371	1.543	1.709	1.871	2.03	2.187	2.34	2.495	2.642
0.6	1.0984	1.1942	1.38	1.556	1.734	1.905	2.078	2.243	2.409	2.573	2.735
0.7	1.099	1.1965	1.388	1.572	1.755	1.935	2.115	2.291	2.46	2.64	2.811
0.8	1.0994	1.1976	1.393	1.584	1.772	1.96	2.148	2.335	2.518	2.7	2.881
0.9	1.0996	1.199	1.397	1.593	1.785	1.984	2.174	2.366	2.561	2.751	2.941
1.0	1.1	1.2	1.4	1.6	1.8	2.0	2.2	2.4	2.6	2.8	3.0
1.1	1.1002	1.2009	1.402	1.606	1.809	2.018	2.22	2.428	2.635	2.842	3.044
1.2	1.1003	1.2015	1.405	1.612	1.821	2.029	2.24	2.45	2.665	2.88	3.09
1.3	1.1004	1.2021	1.408	1.616	1.83	2.039	2.255	2.471	2.697	2.915	3.14
1.4	1.1005	1.2025	1.41	1.621	1.836	2.053	2.271	2.498	2.729	2.946	3.175
1.5	1.1006	1.2032	1.411	1.625	1.842	2.061	2.288	2.513	2.743	2.972	3.205
1.6	1.1007	1.2035	1.414	1.628	1.849	2.07	2.302	2.53	2.765	2.999	3.24
1.7	1.1009	1.2039	1.415	1.632	1.854	2.074	2.311	2.544	2.785	3.028	3.265
1.8	1.101	1.2041	1.416	1.635	1.861	2.085	2.321	2.56	2.799	3.043	3.29
1.9	1.1011	1.2044	1.417	1.637	1.864	2.095	2.33	2.571	2.818	3.067	3.32
2.0	1.1012	1.2049	1.418	1.639	1.865	2.099	2.341	2.585	2.83	3.085	3.342

Table 3 : $Y = \left[1 + 3\left(x^{\frac{1}{a}} - 1\right)\right]^{a}$

a \ X	1.05	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2.0
0.5	1.1434	1.2762	1.5225	1.752	1.97	2.179	2.381	2.582	2.778	2.975	3.16
0.6	1.1456	1.284	1.545	1.794	2.03	2.26	2.49	2.708	2.923	3.142	3.355
0.7	1.471	1.289	1.564	1.827	2.081	2.332	2.58	2.822	3.064	3.3	3.532
0.8	1.1484	1.2935	1.578	1.854	2.128	2.395	2.66	2.923	3.184	3.441	3.7
0.9	1.1492	1.2965	1.589	1.88	2.162	2.454	2.733	3.012	3.298	3.575	3.858
1.0	1.15	1.3	1.6	1.9	2.2	2.5	2.8	3.1	3.4	3.7	4.0
1.1	1.1503	1.302	1.608	1.914	2.231	2.545	2.855	3.175	3.491	3.81	4.135
1.2	1.1511	1.305	1.615	1.931	2.27	2.58	2.905	3.241	3.58	3.905	4.255
1.3	1.1517	1.3065	1.624	1.948	2.281	2.617	2.961	3.305	3.665	4.015	4.375
1.4	1.152	1.308	1.629	1.96	2.304	2.655	3.01	3.372	3.74	4.11	4.49
1.5	1.1522	1.309	1.634	1.972	2.321	2.681	3.052	3.423	3.81	4.2	4.594
1.6	1.1528	1.3105	1.64	1.983	2.341	2.708	3.09	3.476	3.875	4.275	4.685
1.7	1.153	1.312	1.643	1.995	2.359	2.735	3.125	3.523	3.935	4.359	4.775
1.8	1.1533	1.3135	1.6475	2.004	2.378	2.755	3.155	3.572	3.995	4.42	4.868
1.9	1.1534	1.3137	1.651	2.01	2.39	2.785	3.14	3.612	4.05	4.499	4.945
2.0	1.1537	1.314	1.656	2.016	2.398	2.795	3.222	3.654	4.11	4.555	5.04

(Table 2)
(Table 3)

in the charts and the tables, the larger the values of X and Y, the better estimation of α we may get. When the value of α is estimated, the corresponding value of t_o may be obtained from the equation (39) or (40) to be as follows;

$$t_o = t - \frac{\Delta t}{\left(\frac{I}{I_1}\right)^{\frac{1}{\alpha}} - 1} \qquad (43)$$

or

$$t_o = t - \frac{k\Delta t}{\left(\frac{I}{I_k}\right)^{\frac{1}{\alpha}} - 1} \qquad (44)$$

where t is the time of the first measurement of the activity I, and I_1 and I_k are the activities after the time interval Δt and $k\Delta t$ from the time t of the first measurement respectively.

However, in order to facilitate the rough order of magnitude estimation of $\iota = t - t_o$, the time from the nuclear detonation to the time of the first measurement of the activity in the triplet set of measurements used for the estimation, another chart as shown in Fig.4 was constructed based upon the following equation corresponding to the equation (39).

$$X^{\frac{1}{\alpha}} - 1 = Z \qquad (45)$$

where $X = \frac{I}{I_1}$ and $Z = \frac{\Delta t}{t - t_o}$.

When we read off Z for the corresponding values of X and α, the time ι of the nuclear detonation prior to the first measurement may easily be estimated from the following simple relation.

$$\iota = t - t_o = \frac{\Delta t}{Z} \qquad (46)$$

The numerical values, based on which the chart in Fig.4 was contructed, are given in Table 4. The above charts and tables were computed for the ranges of X from 1.05 to 2.0 and α from 0.5 to 2.0 which are expetected to cover practically most of the cases according to our past experience.

(Fig. 4)

(Table 4)

With the above method using the charts or tables, we may estimate the values of t_o and α. However, the different triplet set of measurements may sometimes give a considerably different values of t_o and α especially when the statistical fluctution of counting the activity is not sufficiently small.
In such cases it may be safer to try to obtain the most probable estimates of t_o and α from different triplet sets of measurements extracted from a series of measurements for a sufficient length of time.

However, in case the most probable estimate of α is obtained first, we may estimate t_o graphically by plotting $I_i^{-\frac{1}{\alpha}}$ against t_i and obtaining the value of t corresponding to $I_i^{-\frac{1}{\alpha}} = 0$ by extrapolating the straight line according to the equation.

$$I_i^{-\frac{1}{\alpha}} = A_o^{-\frac{1}{\alpha}} (t_i - t_o) \qquad (47)$$

In this case, the same analytical method as derived by the equations (16) - (20) in the Method (I) may be applicable in estimating the values of A_o and t_o from a series of n measurements (I_i, t_i), (i = 1,2, ------, n)

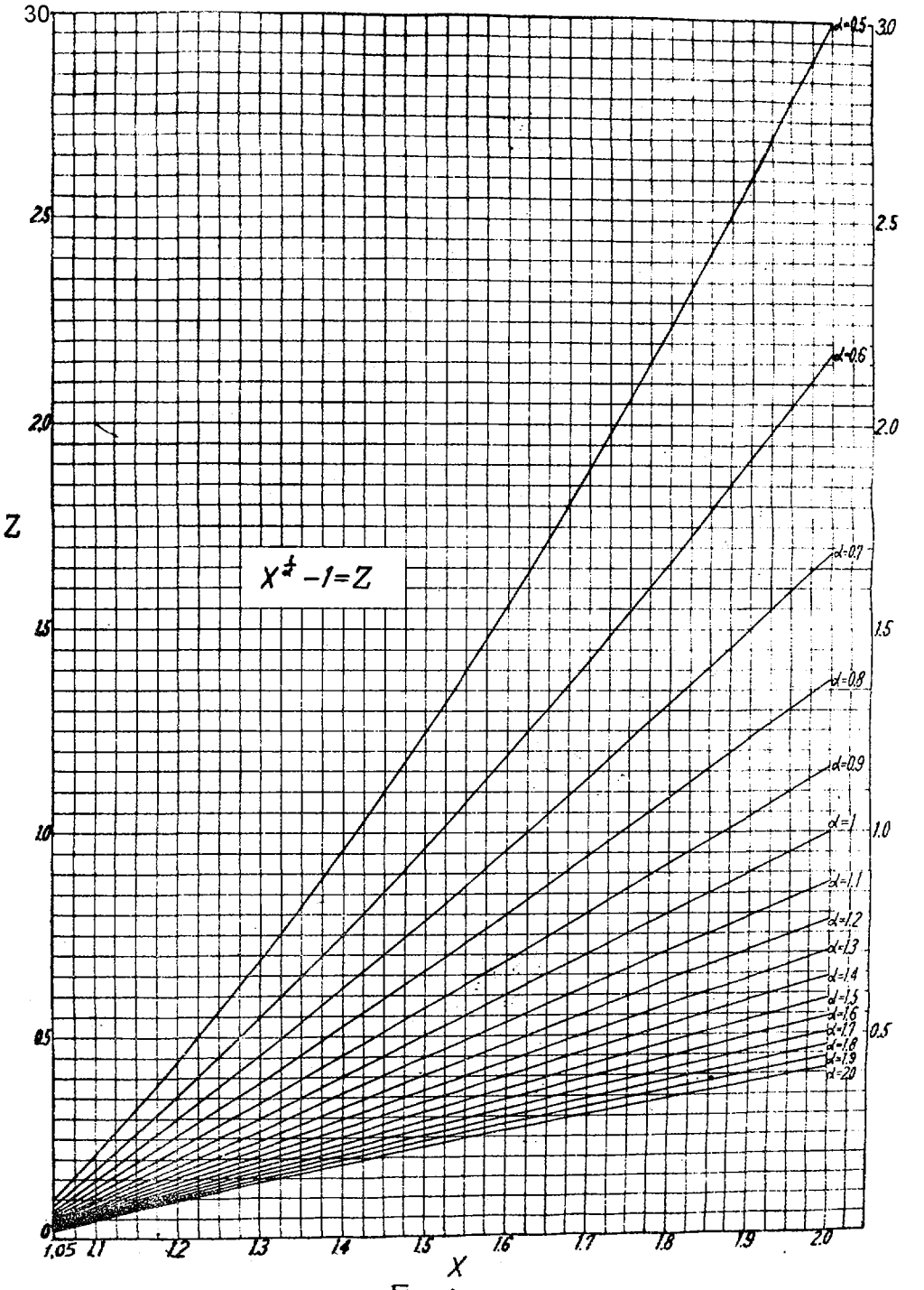

30
25
20
Z
15
10
0.5
0
$X^{\alpha} - 1 = Z$
α=0.5
α=0.6
α=0.7
α=0.8
α=0.9
α=1
α=1.1
α=1.2
α=1.3
α=1.4
α=1.5
α=1.6
α=1.7
α=1.8
α=1.9
α=2.0
3.0
2.5
2.0
1.5
1.0
0.5
1.05
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2.0
X

Fig. 4

Table 4 : $Z = X^{\frac{1}{\alpha}} - 1$

α \ X	1.05	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2.0
0.5	0.1025	0.21	0.44	0.69	0.96	1.25	1.56	1.89	2.24	2.61	3
0.6	0.0847	0.172	0.355	0.548	0.751	0.9655	1.19	1.42	1.663	1.915	2.175
0.7	0.0722	0.146	0.298	0.455	0.617	0.785	0.957	1.134	1.32	1.5	1.69
0.8	0.0629	0.1265	0.256	0.388	0.523	0.66	0.8	0.941	1.085	1.23	1.378
0.9	0.0556	0.1117	0.225	0.339	0.452	0.57	0.685	0.802	0.922	1.04	1.16
1.0	0.05	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.
1.1	0.0454	0.0905	0.18	0.269	0.358	0.446	0.533	0.62	0.706	0.792	0.878
1.2	0.0415	0.0827	0.164	0.244	0.324	0.401	0.479	0.555	0.632	0.706	0.781
1.3	0.0382	0.0761	0.1505	0.224	0.296	0.365	0.435	0.504	0.572	0.638	0.705
1.4	0.0354	0.0704	0.139	0.206	0.272	0.336	0.399	0.461	0.522	0.582	0.641
1.5	0.033	0.0657	0.1291	0.191	0.252	0.31	0.368	0.424	0.48	0.534	0.5875
1.6	0.0309	0.0614	0.1208	0.178	0.234	0.288	0.342	0.393	0.444	0.493	0.542
1.7	0.0291	0.0576	0.1131	0.167	0.219	0.269	0.3185	0.366	0.413	0.459	0.503
1.8	0.02745	0.05434	0.1066	0.157	0.206	0.252	0.298	0.343	0.386	0.428	0.47
1.9	0.026	0.0514	0.1007	0.148	0.194	0.238	0.2805	0.322	0.3625	0.402	0.44
2.0	0.0247	0.0488	0.0955	0.14	0.183	0.224	0.265	0.304	0.3419	0.3785	0.414

Method IV

This method may be called a differential method. Differentiating the following relation between I and t,

$$I = A_0(t - t_0)^{-\alpha(t)} \tag{48}$$

with respect to t, we may obtain in general the expression.

$$\frac{1}{I}\frac{dI}{dt} = -\frac{d\alpha(t)}{dt}\ln(t - t_0) - \alpha(t)\frac{1}{t - t_0} \tag{49}$$

If we assume $\alpha(t) = \alpha$ is a constant for a certain time interval as before, we may obtain

$$-\frac{1}{I}\frac{dI}{dt} = \alpha\frac{1}{t-t_0} \tag{50}$$

As clear from tne above equation, if we put

$$Y = 1 \Big/ \left(-\frac{1}{I}\frac{dI}{dt}\right) \tag{51}$$

the following linear relation may be obtained between Y and t,

$$Y = \frac{1}{\alpha}(t - t_0) \tag{52}$$

Therefore, by plottling the estimated Y against t extrapolating the straight line to a point corresponding to $Y = 0$, we may estimate t_0 graphically and as a reciprocal of the slope of the line the value of α may also be estimated.

$$\alpha = \frac{t-t_0}{Y} \tag{53}$$

Terasaki[5] used this graphical method in estimating t_0.

However, as pointed out before, a more objective estimation may be obtained by the following analytical method.

Assuming α and t_o are constants in the equation (52), for a series of estimated values (Y_i, t_i), $(i = 1, 2, \text{------}, n)$, the error may be expressed by

$$S = \Sigma\left[Y_i - \frac{1}{\alpha}(t_i - t_o)\right]^2 \qquad (54)$$

For minimum error in the estimation of the constants t_o and α,

$$\frac{\partial S}{\partial t_o} = 0, \quad \frac{\partial S}{\partial \alpha} = 0 \qquad (55)$$

Solving the above equations, t_o and α may be estimated to be as follows.

$$t_o = \frac{\Sigma Y_i \ \Sigma(t_i)^2 - \Sigma Y_i t_i \Sigma t_i}{\Sigma Y_i \Sigma t_i - n\Sigma Y_i t_i} \qquad (56)$$

$$= \frac{-(\Sigma t_i)^2 - n\Sigma(t_i)^2}{\Sigma Y_i \Sigma t_i - n\Sigma Y_i t_i} \qquad (57)$$

where

$$Y_i = 1 \Big/ \left(-\frac{1}{I}\frac{dI}{dt}\right)_{t=t_i} \qquad (58)$$

If the time interval between the two successive measurements is small and the deviation from the Way-Wigner relation or the fluctuation of the activity is small, the value of Y corresponding to the time t may be approximately estimated from the two successive measurements (I_i, t_i) and (I_{i+1}, t_{i+1}) as follows:

$$Y = \frac{1}{2}(I_i + I_{i+1})\frac{(t_{i+1} - t_i)}{(I_i - I_{i+1})} \qquad (59)$$

corresponding to the time t given by

$$t = \frac{t_i + t_{i+1}}{2} \qquad (60)$$

However, according to the author's experience, a better result seem to be obtained if $Y = 1/\left(-\frac{1}{I}\frac{dI}{dt}\right)$ is properly estimated from a properly smoothed out decay curve.

In the foregoing methods α has been assumed to be constant. However, an attemp was made by a study group on the effects of A and H bomb in the Meteorological Research Institute[6] to try to estimate t_o assuming α is a function of t. In this method $\alpha_{(t)}$ is assuming to change with time in the same way as the composite activity given by Hunter and Ballou[7]. Sugiura[8] pointed out that this method seems to be rather difficult unless the composition of the radioactive nuclides is exactly the same as given by Hunter and Ballou. Since the composition of the various nuclides in the sample may not be considered always the same especially in the case of detection of activity at a remote place, there seems to be a danger that a more erroneous result might be obtained with this method than with the method assuming the Way-Wigner relation with constant α for a reasonable period of time. Although it was noticed by the author that the value of α changes occasionally after a certain period of time, if α should change drastically beyond the limit of experimental error, it would be noticed during the analysis with the assumption of constant α. Therefore, for practical purposes, the method assuming the Way-Wigner relation with constant α seems to be quite satisfactory in order to obtain the order of magnitude estimation of t_o and α, unless the type of the nuclear detonation drastically changes in the future.

Method V

This method may be called an integral method. Assuming the Way-Wigner relation with constant α, we may have the following expression.

$$I = A_o (\tau_o + t)^{-\alpha} \qquad (61)$$

where τ_o is the time from the nuclear detonation to the time of the first measurement of the activity, and t the time from the first measurement.

Integrating the equation (61) with respect to t for the integral limit from t = 0 to t = t_1, the total activity $(TA)_{t_1}$ as measured during the time interval t_1 may be expressed by the following equation

$$(TA)_{t_1} = \int_o^{t_o} I\, dt = \int_0^{t_1} A_o (\tau_o + t)^{-\alpha} dt \qquad (62)$$

Performing the integration, we may obtain

$$(TA)_{t_1} = \frac{1}{1-\alpha} \left[I_1 (\tau_o + t_1) - I_o \tau_o \right] \qquad (63)$$

where I_o is the activity at t = 0 or at the first measurement which was conducted at the time τ_o after the nuclear detonation and may be expressed by,

$$I_o = A_o \tau_o^{-\alpha} \qquad (64)$$

and I_1 is the activity at the time $t = t_1$ after the first measurement or at the time $(\tau_o + t_1)$ after the nuclear detonation and may be expressed by,

$$I_1 = A_o (\tau_o + t_1)^{-\alpha} \qquad (65)$$

Similarly, the total activity $(TA)_{t_2}$ as measured during the time interval t_2 since the beginning of the first measurement may be expressed by the following equation.

$$(TA)_{t_2} = \int_o^{t_2} I\, dt = \int_0^{t_2} A_o (\tau_o + t)^{-\alpha} dt \qquad (66)$$

Performing the integration, we may obtain,

$$(TA)_{t_2} = \frac{1}{1-\alpha}\left\{ I_2(t_o + t_2) - I_o t_o \right\} \quad (67)$$

where I_o is the activity at the first measurement

$$I_o = A_o\ t_o^{-\alpha} \quad (68)$$

and I_2 the activity at the time t_2 from the first measurement

$$I_2 = A_o\ (t_o + t_2)^{-\alpha} \quad (69)$$

Dividing the equation (67) by the equation (63),

$$\frac{(TA)_{t_2}}{(TA)_{t_1}} = \frac{I_2(t_o + t_2) - I_o t_o}{I_1(t_o + t_1) - I_o t_o} \quad (70)$$

Solving the above equation with respect to t, we may obtain,

$$t_o = \frac{(TA)_{t_2}\ t_1 I_1 - (TA)_{t_1} t_2 I_2}{(TA)_{t_2}(I_o - I_1) - (TA)_{t_1}(I_o - I_2)} \quad (71)$$

Substituting the above value of t_o into equation (63) or (67), and solving for α, we may obtain,

$$\alpha = \frac{(I_o - I_1)\left\{(TA)_{t_2} - t_2 I_2\right\} - (I_o - I_2)\left\{(TA)_{t_1} - t_1 I_1\right\}}{(TA)_{t_2}(I_o - I_1) - (TA)_{t_1}(I_o - I_2)} \quad (72)$$

where $(TA)_{t_1}$ and $(TA)_{t_2}$ are the total activities for the time intervals from $t = 0$ to $t = t_1$ and $t = t_2$ respectiverly, and I_o, I_1 and I_2 are the activities measured at $t = 0$, $t = t_1$ and $t = t_2$ respectively, and $t = 0$ is the beginning of the first measurement which is started at the time τ_o after the nuclear detonation.

From the series of measurements which are started at the different times and for the different time intervals, we may obtain somewhat different values of t_o and α. Therefore, in this case also, it would be better to estimate the most probable values of t_o and α as a mean value by treating statistically somewhat different values estimated from different series of measurements.

However, since this method involves the total activities for the period of t_1 and t_2, the fluctuation of activity may be more or less cancelled out during the continuous measurement. In this respect, this integral method, if properly applied, seems to be better as compared with other methods which depend on interpolation or extrapolation.

Method VI

Chatterjee reported the following method as a method of dating a nuclear explosion.

According to Way and Wigner the composite beta-decay resulting from nuclear fission can be represented, using the notations used in this paper, as

$$I \propto \tau^{-\alpha} \qquad (73)$$

where I is the activity at time τ days after explosion and α is a constant. The theoretical value of α has been given to be 1.2, while the experimental value varies from 0.8 to 1.9. Holter and Glasscock have found that for nuclear fission bomb, the same relation holds with $\alpha = 1.3$. The Way-Wigner relation can be used to verify whether the radioactivity of the dusts is due to the decay of fission products.

It is assumed, a priori, that the Way- Wigner law holds for the composite beta-activity resulting from the thermonuclear fusion bomb, the value of α will have to be found.

The experiment consists in measurement of the activity I_1 of a given sample at a time τ_1 after explosion. The dusts are collected on a particular day and the radioactivity on the same day ($t = 0$) is I_0.

On subsequent days ($t = t_1, t_2, t_3, \text{------}t_i, \text{------}tn$) the corresponding measures of radioactivity are

$$(I_1, I_2, I_3, \text{------}I_i, \text{------}I_n).$$

Suppose that the explosion has taken place τ_o days before the first measurement of activity. Then,

$$\tau_o = \tau_o + 0,\ \tau_1 = \tau_o + t_1,\ \tau_2 = \tau_o + t_2,\ \text{------}\tau_i = \tau_o + t_i,\text{------}\ \tau_n = \tau_o + t_n. \tag{74}$$

From the results of the decay measurements we have a set of data:

$$(I_o, 0), (I_1, t_1), (I_2, t_2), \text{------}(I_i, t_i), \text{------}(I_n, t_n), \tag{75}$$

for n + 1 observations. From equation (73) we find

$$I_i^{-\frac{1}{\alpha}} = k\tau_i = k(\tau_o + t_i) \tag{76}$$

where k is a constant. If we assume the value of α from Way-Wigner, the $I_i^{-\frac{1}{\alpha}}$ vs t_i plot will be a straight line the extrapolation of which will give the value of τ_o.

However α and τ_o can be found separately if the $I \propto \tau^{-\alpha}$ law holds. The experimental error can be expressed as

$$S = \Sigma\left(\ln\frac{I_i}{I_o} + \alpha \ln\frac{\tau_i}{\tau_o}\right)^2 \tag{77}$$

S involves the errors in estimating α and τ_o. For minimum error in α and τ_o, $\frac{\partial S}{\partial \alpha} = 0$ and $\frac{\partial S}{\partial \tau_o} = 0$, from which we obtain,

$$\alpha = \alpha(\tau_o) = \frac{\Sigma \ln\frac{I_o}{I_i} \ln\frac{\tau_i}{\tau_o}}{\Sigma\left(\ln\frac{\tau_i}{\tau_o}\right)^2} \tag{78}$$

and

$$\alpha = \alpha'(\tau_o) = \frac{\Sigma\left(\frac{1}{\tau_o} - \frac{1}{\tau_i}\right)\ln\frac{I_o}{I_i}}{\Sigma\left(\frac{1}{\tau_o} - \frac{1}{\tau_i}\right)\ln\frac{\tau_i}{\tau_o}} \tag{79}$$

where $\tau_o = \tau_o + t_i$.

For any assumed value of τ, the above equations will

give the mean value of α,

$$\alpha = \frac{\alpha(\iota_o) + \alpha'(\iota_o)}{Z} \qquad (80)$$

For correct value of α and ι_o, S of equation (77) will be minimum. From equation (76) an idea about the probable date of explosion or nuclear detonation can be estimated. Then for different values of ι_o around the probable value, the mean value of α and the corresponding value of ι can be calculated. From the $S_{(\iota_o)}$ vs ι_o plot, the actual value of ι_o can be obtained for minimum value of $S_{(\iota_o)}$. The resolution of the $S_{(\iota_o)}$ vs ι_o plot will also indicate the errors involed in finding α and ι_o.

Although Chatterjee assumed the value of $\alpha = 1.3$ in estimating the approximate value of ι_o graphically according to the equation (76), the above analytical method may be applicable to finding the most probable value of ι_o when the approximate values of ι_o and α are estimated by other methods described previously in this paper.

Chatterjee estimated, from the analysis of the radioactivity of the samples collected at Calcutta in India in the second half of May, 1954, that an unannounced nuclear detonation appeared to have been conducted around May 12.

Although the analysis was made independently with somewhat different method by the Japanese scientists,[4][10] a similar result was obtained from the analysis of the radioactivity of the samples collected in Japan in the second half of May, 1954, that an unannouced nuclear detonation seems to have been

conducted during the period from May 10 to May 13, 1954. It should be noted that the same result was also obtained from the analysis of the radioactivity of the samples collected from the Japanese boats which were containated while passing outside the Waring Area in the South Pacific during the period from May 13, 1954.(4)

(II) Discussion and Summary

In this paper various methods of estimating the probable time of nuclear detonation are introduced. However, since all of the methods described in this paper have been derived based upon the assumption that the Way-Wigner relation $I^{\infty} t^{-\alpha}$ holds with constant α, one method may be considered a modification of another.

(Fig.5)

(Fig.6)

Some of the results of the analysis with the different methods are given in Figs. 5 - 11. However, these are the examples of the analysis with which a considerably good approximation seemed to have been obtain. Usually, the error of a few days seems to be unavoidable in estimating the probable time of nuclear detonation from the analysis of the grossactivity with these method assuming the Way-Wigner relation with constant α. Fig.5 is an example of the analysis of the radioactivity which could be ascribed to March 1 test in 1954. Fig.6 shows the results of the analysis with the Method (II) of the samples collected in Osaka from the middle to the second half of May, 1954. As can be seen in the figure, the radioactivity collected from May 14 rain and that from May 17 rain appeared to be due to different nuclear detonation at different date.

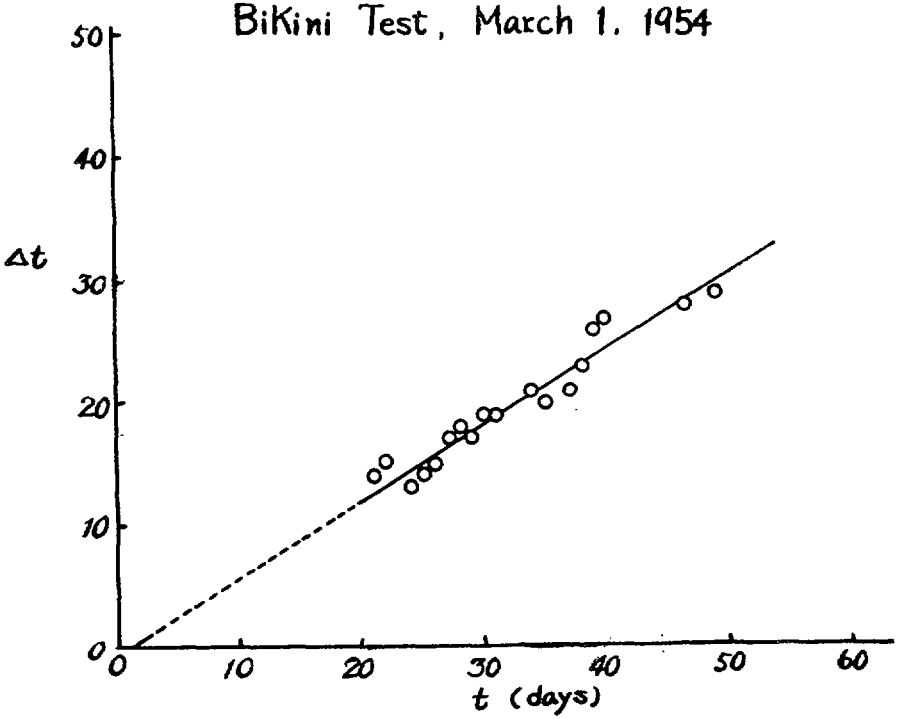

BiKini Test, March 1. 1954
Δt
50
40
30
20
10
0
0
10
20
30
40
50
60
t (days)

Fig. 5

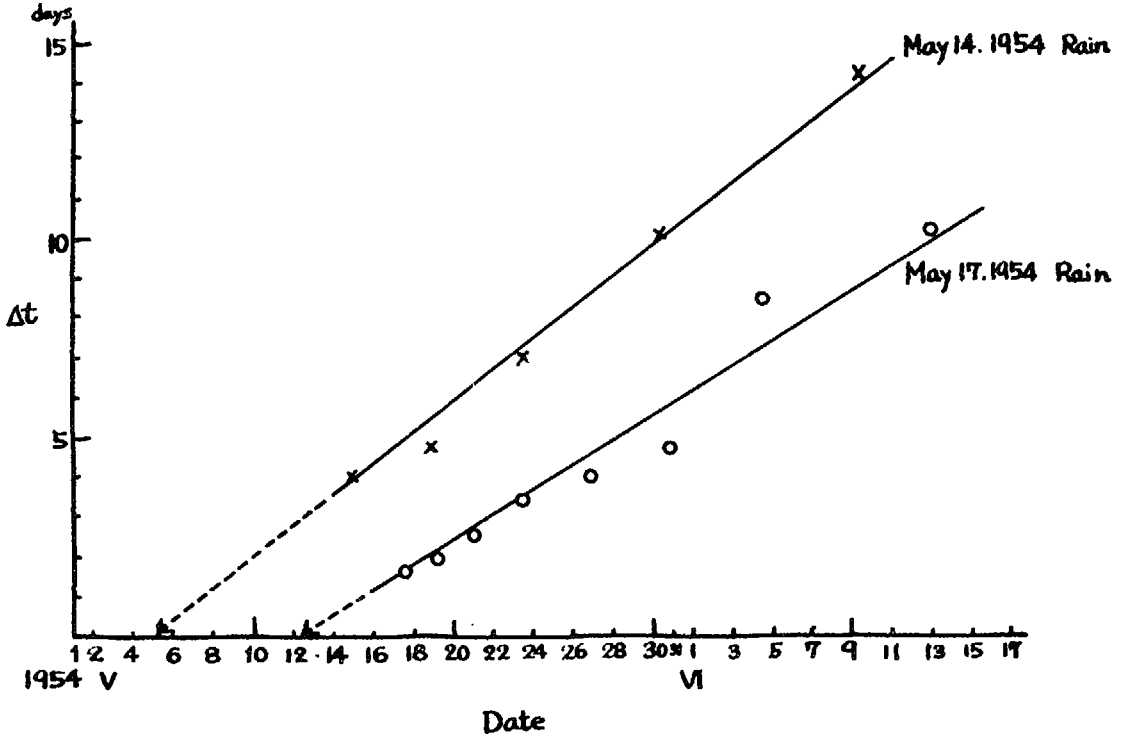

days
15
10
5
Δt
May 14. 1954 Rain
May 17. 1954 Rain
1 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30 31 1 3 5 7 9 11 13 15 17
1954 V
VI
Date

Fig. 6

Although the result of this particular analysis of May 17 sample indicates that the radioactivity in the sample might be due to a nuclear detonation around May 12, the probable time of detonation estimated with the Method (I) and Method (V) was observed to fall in the range between May 8 to May 13.
The radioactive decay corresponding to Fig.6 is plotted in full logarithmic scale in Fig.7, assuming the estimated date as the probable time of detonation. Fig.8 is an example of the analysis of May 14 sample with the Method (IV). In the figure the cross sign indicates the point obtained by the approximation according to the equations (59) and (60) based upon the directly measured values, while the circle indicates the point estimated from a smoothed out curve.

(Fig.7)
(Fig.8)

As can be seen in this example, if the interval between the two successive points of measurements is not small enough, the estimated points according to the equations (59) and (60) may be scatterd around considerably. Even the points estimated from the smoothed out decay curve are usually scattered about a little more than this with method, because of the difficulty of estimating the tangent at a point on the curve accurately.

For the purpose of comparison, the results of the analysis with the Method IV by Chatterjee[9] on the samples collected at Calcutta in the second half of May in 1954 are shown in Fig.9. As can be seen in the figure, his results seem to agree well with the result of our analysis on May 17 rain in Osaka which is shown in Fig.6 although the value of α seems to be somewhat different.

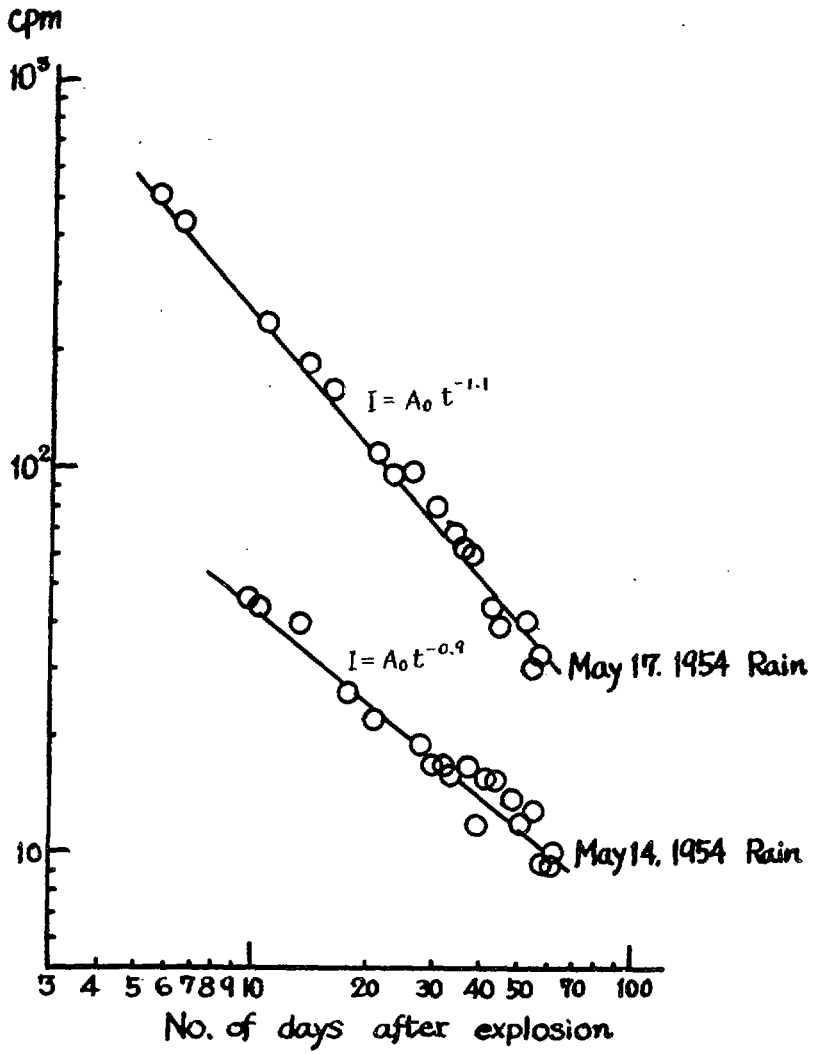

cpm
10^3
10^2
10
$I = A_0 t^{-1.1}$
$I = A_0 t^{-0.9}$
May 17. 1954 Rain
May 14. 1954 Rain
3 4 5 6 7 8 9 10 20 30 40 50 70 100
No. of days after explosion

Fig. 7

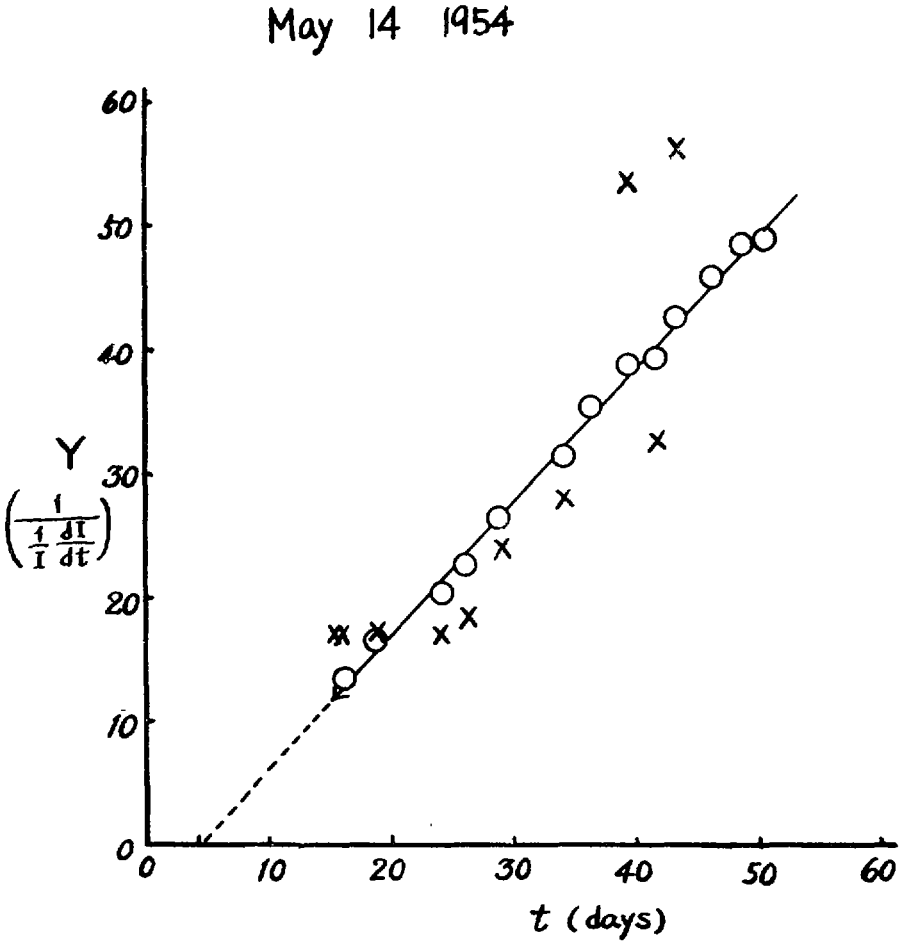

May 14 1954
60
50
40
30
20
10
0
Y
$\left(\frac{1}{\frac{1}{I}\frac{dI}{dt}}\right)$
0
10
20
30
40
50
60
t (days)

Fig. 8

(Fig.9)
(Fig.10)
(Fig.11)

Fig.10 shows the result of the analysis with the Method (III) on the samples collected from March 1 rain in 1960. With the method of analysis using the charts shown in Figs.2 and 3, if we try to estimate τ_0 and α using only one triplet points of actual measurements with small values of X, Y and Δt, the error in the estimation was observed to be so large that the value of α ranged from 0.6 to 1.9 and the error of the order of 5 - 10 days in the estimation of the probable date of detonation was not seldom. Therefore, we have tried to estimate a number of values of τ_0 and α by shifting the original point with the value of Δt abput 5 - 10 days. By treating these somewhat different values of τ_0 and α statistically, we have obtained the most probable value of $\alpha = 1.4$ and the corresponding value of $\tau_0 = 20$ days prior to March 5. Assuming these values were correct, an example of $I_i^{-\frac{1}{\alpha}}$ vs τ_i plot based upon the actually measured values is given in Fig.10, and the corresponding I_i vs τ_i plot in full logarithmic scale is shown in Fig.11. The points in Fig.10 may be observed to be well approximated by a stright line the extrapolation of which can be seen to fall around Feb.13 or in the range from Feb.12 - 15. Since the only announced nuclear explosion test during this period is the French test conducted in Sahara desert on Feb.13, 1960, the corelation between the estimated probable time of detonation and the French test in Sahara seems to be clear. Although the Method(III) was developed in the hope of estimating the probable date of nuclear detonation based upon the actually measured values rapidly and more objectively without depending upon extrapolation or interpolation from the arbitrarily smoothed out curves, the probable time of nuclear detonation estimated with this particular example

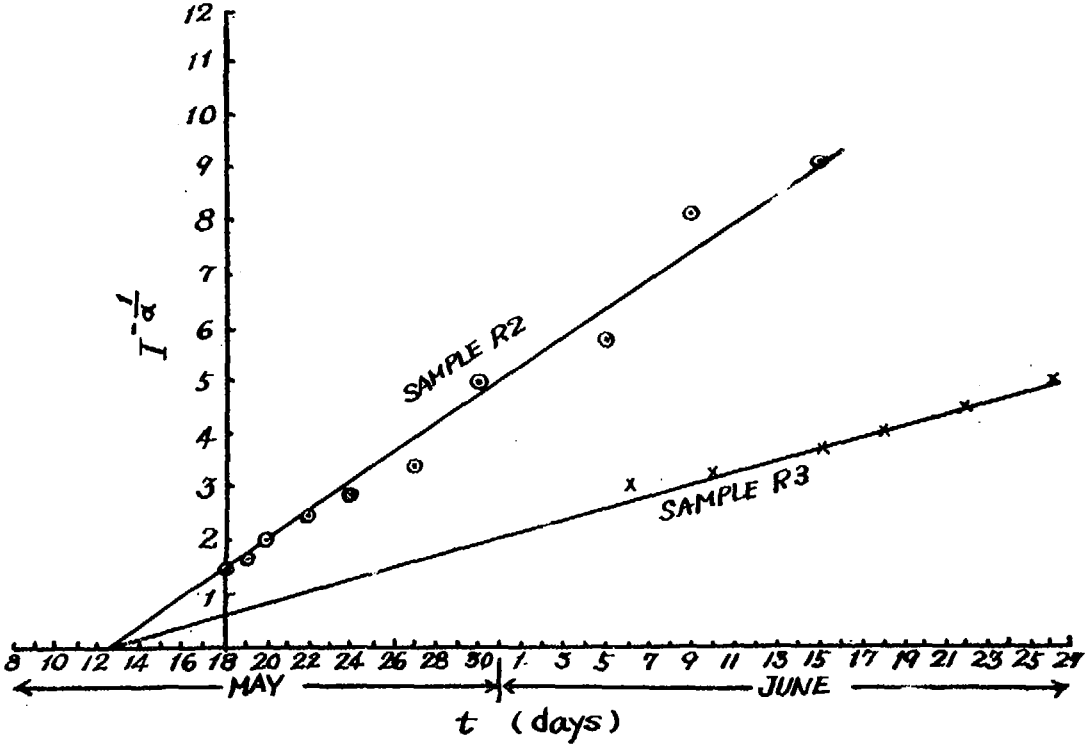


Fig. 9: Tracing back the date of explosion from the Way-Wigner relation ($\alpha = 1.3$) (after Chatterjee)

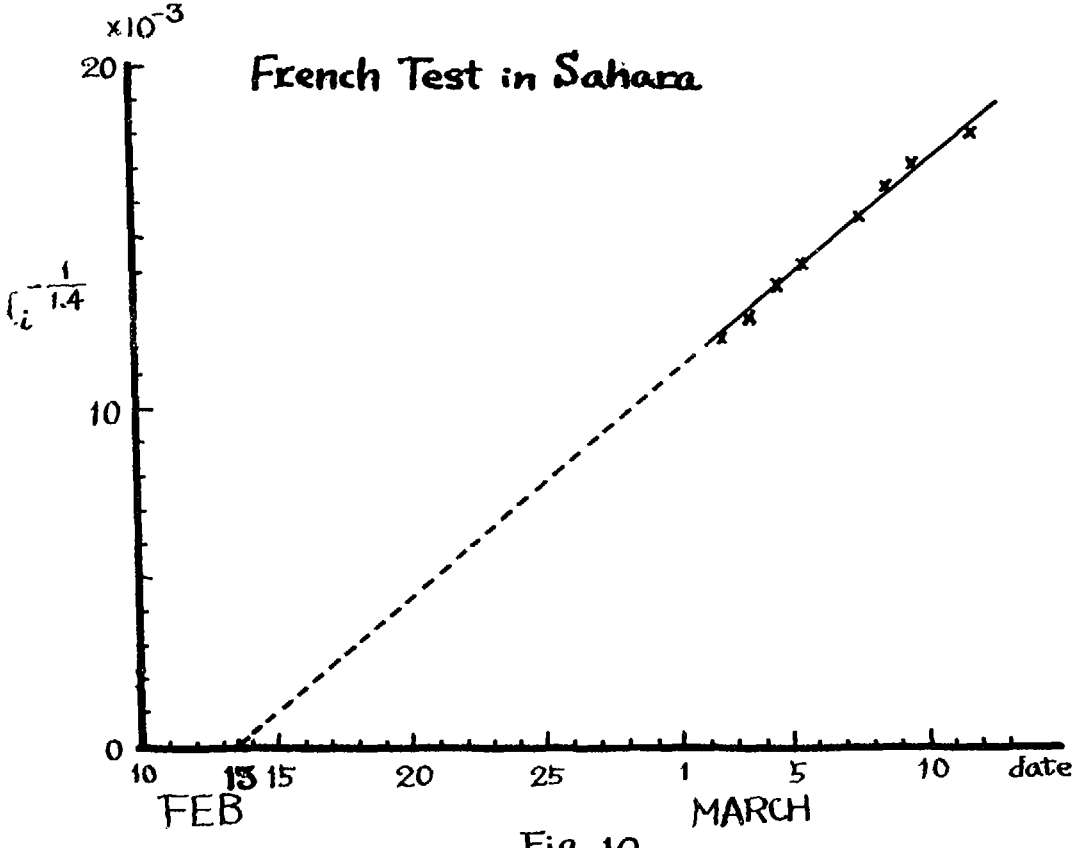

French Test in Sahara
$\times 10^{-3}$
20
10
0
$\left(\frac{1}{i}\right)^{\frac{1}{1.4}}$
10
15
15
20
25
1
5
10
date
FEB
MARCH

Fig. 10

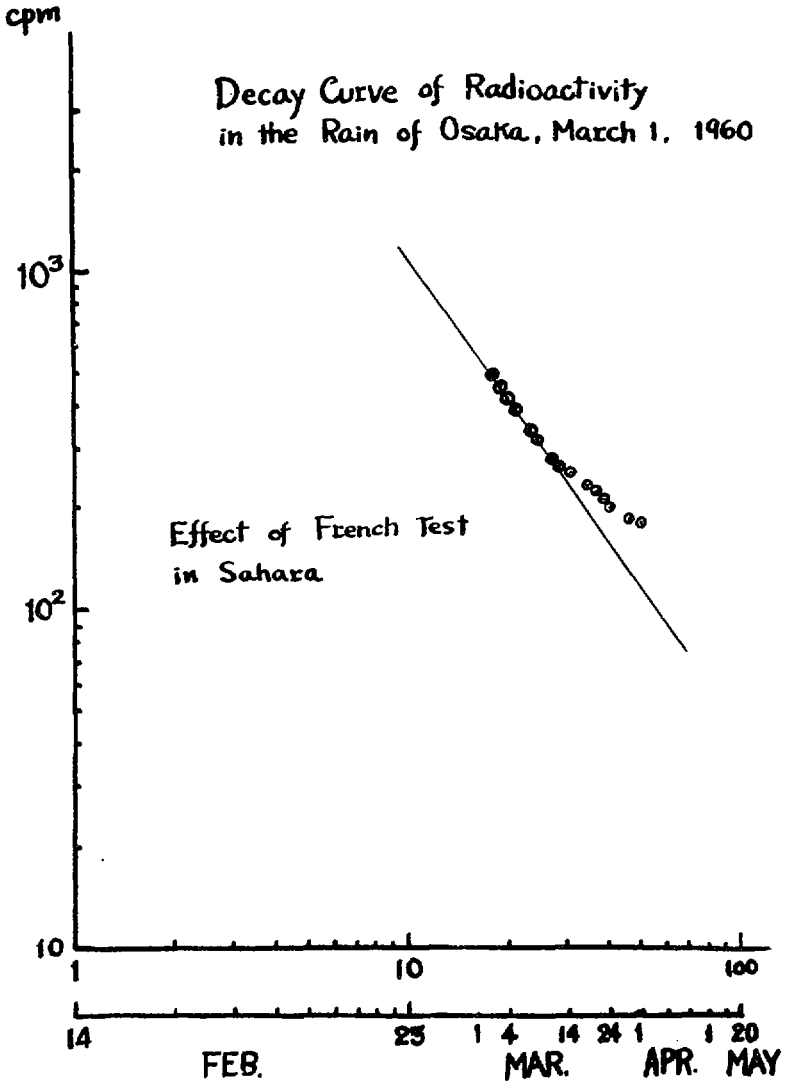

cpm
Decay Curve of Radioactivity
in the Rain of Osaka, March 1, 1960
10^3
10^2
10
Effect of French Test
in Sahara
1
10
100
14
25
1
4
14
24
1
1
20
FEB.
MAR.
APR.
MAY
1960

Fig. 11

based upon only one set of triplet measurements with small values of X, Y and Δt was observed to lie in such a wide range as from Feb.5 to about Feb.25.

As shown in this example, the error in the estimation of t_o with the various methods described in this paper will in general be expected to be considerably large if we try to estimate the probable time of nuclear detonation from a limited small number of measurements or extrapolated or interpolated points unless those those points are the ones carefully selected from the very properly smoothed out decay curve. Therefore, usually, it seems to be necessary to try to estimate the most probable values from a number of measurements or extrapolated or interpolated points for a sufficient and adequate time interval, during which the value of α of the Way-Wigner relation may be considered approximately constant, in order to obtain a better result of estimation.

Although the value of α, as observed at a remote place such as Japan, was observed to change during the period of about one to two months after the nuclear detonation, if the methods described in this paper were properly applied, it may be possible to test whether the assumption of Way-Wigner relation with constant α would hold for a long period of observation.

Acknowledgement

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Studies on the Radioactive Contamination due to Nuclear Detonations IV

(On the Artificial Radioactivity in the Rain of Osaka during the period from April 1954 to the end of August 1961)

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On the Artificial Radioactivity in the rain of Osaka during the Period from April 1954 to the end of August 1961.

The results of the routine measurements of the radioactive fallout as observed in Osaka, Japan during the period from 1954 to the end of August 1961 are introduced in this paper.

(I) Method of Measurement

The rainwater was collected by the polyethylene vessels with the effective surface area for collection of rainwater about 1square meter. The smaller vessel was used for the daily routine measurements of the grossactivity of the radioactive fallout and the larger one was kept for one year for the estimation of the annual precipitation of strontium-90 and cesium-137.

The gross beta activity of each rainfall was measured with the residue obtained after complete evaporation of an aliquot of the rainwater by the commercial beta ray counter with a thin mica window of thickness about 1.9 mg/cm^2 manufactured by Kobe Kogyo K.K..

In order to avoid the possible complications due to the relatively short lived naturally occurring disintegration products of Rn or Tn, the grossactivity at the time of sampling was estimated by the back extrapolation based upon the decay rate from sereral hours to a few days after sampling.

The dry deposition was also measured by exposing a greased paper with the surface area about 0.1 square meter to the air for a certain period of time. The collected radioactive dust was wrapped up in the paper and completely ashed in the electric furnace. After weighing the ash, the gross beta activity was measured more than 24 hours after sampling. When the amount of

ash is large, a correction for the self-absorption due to sample thickness was attempted.

The radioactivity in unit of curies was estimated from the comparison with the substandard source of Sr^{90} + Y^{90} which was previously calibrated by RaE. For the accurate measurements of radioactivity 4π-counter which was specially manufactured for our laboratory by Kobe Kogyo K.K. was used. From the comparative measurements of the artificial radioactivity in the dust collected from the rainwater sample with the 4π-counter and with the usual beta ray counter, the efficiency of counting the beta rays of the sample by the beta ray counter at the usual geometry used for routine measurements was estimated to be about 7.5%, while the efficiency ~~of counting~~ of counting high energy beta rays from Y^{90} was over 10%.

When more accurate estimation of activity is necessary, a low background counter constructed in our laboratory was used. The beta ray counter was surrounded by 22 cosmic ray counters which were constructed at the cosmic ray laboratory of the department of physics of Osaka City University and kindly provided to the author by courtesy of Prof. Watase and Dr. Miyake. The anticoincidence circuit used for the low background counting was constructed by Dr. Kawai of our laboratory. The whole set was surrounded by a heavy shielding of iron sheets which were kindly provided to the author from the Fujinagata Shipbuilding Co. in Osaka by the kind arrangement of Prof. Okuda of the department of physics.

The analysis of strontium-90 and cesium-137 was attempted with the assistance of Mrs. Kuriyama, Mr. Tanaka, Dr. Iwasaki and Mrs. Furukubo and with the kind help by Prof. Hara of

Doshisha University, Prof. Yamatera of Osaka City University and Prof. Nishi of Kyoto University.

The procedures of the analysis of annual sample may be introduced in the following.

1) Take an Aliquot of the rainwater sample from the large rainwater collecting vessel. Add about 50mg each of Sr and Cs carrier, and concentrate by boiling.

2) Evaporate the concentrated solution to complete dryness. Then the residue is ashed at about 600°C in the electric furnace for one two days.

3) Dissolve the residue with 1 - 3N hydrochloric acid and filter.

4) To the filtrate, add ammonium hydroxide solution slowly until a faint turbidity is observed. Add a neutral solution of ferric chloride to the extent that the yellowish colour could be recognized. Then add a little glacial acetic acid, and a concentrated solution of ammonium acetate. Boil the solution for some minutes to precipitate phosphate and filter hot.

The precipitate is redissolved in the minimum quantity of dilute hydrochloric acid and repeat the precipitation to recover strontium in the filtrate.

5) To the combined filtrates, add ammonium chloride solution and ammonium hydroxide solution and boil. (Al, Fe, Cr and rare eath group may precipitate) Pass hydrogen sulphide, boil until the precipitation is complete, and filter. (Zn, Mn, Co precipitate).

6) Concentrate the filtrate. Make the solution acid with acetic acid and drive out H_2S by heating. Make the filtrate neutral to basic with ammonium hydroxide and add a solution of ammonium carbonate (10 - 20g) in excess and 15ml of 95% ethanol. After stirring the solution strongly, digest with heating and allow to stand still more than 30 min. and filter. The precipitate is washed with the solution of ammpnium carbonate to which alcohol is added.

The precipitate is dried at 100 - 110°C and weighed. (Ca, Sr) To the filtrate and wash, add about 0.5 gram of Ca^{++} carrier and repeat the precipitation to recover stronium. These precipitates are combined and reserved for the analysis strontium.

7) The filtrate is evaporated to dryness and the residue is heated at about 500°C for one to two days in the electric furnace to drive out ammonium.

8) Dissolve the residue with water and dilute hydrochloric acid. Concentrate the solution as much as possible, and add a freshly prepared solution of sodium cobaltinitrite $Na_3Co(NO_2)_6$ in excess. Shake vigorously for some minutes or stir mechanically for 30 min. and allow to stand over night. Centrifuge and filter. Wash the precipitate with 10% acetic acid and then with 95% alcohol. The precipitate is dried at 100 - 110°C and weighed. (K, Cs)

9) The reagent of sodium cobaltinitrite may be prepared as follows: Dissolve 28.6g of cobaltous nitrate crystals and 50ml of glacial acetic acid in enough water to make 500ml of solution. Dissolve 180g of sodium nitrite in 500ml of water. Keep these two solutions separate until the day before analysis, then mix equal volumes, shake or stir well, allow to stand over night in a glass-stoppered bottle, and filter just before using.

10) The precipitate in step (8) is mixed with a little sodium nitrite and heated until the mass fuses and effervesces no more. The residue is taken up in warm water and filter off the black cobalt oxide residue after washing with water thoroughly. To the combined solution of the filtrate and the wash, add hydrochloric acid and boil to decompose the nitrite and evaporate to dryness to eliminate nitrous acid completely.

11) Dissolve the residue in warm water and make acid with glacial acetic acid and then add about 5ml of $HBiI_4$ reagent. Cool the mixture with ice and centrifuge. Wash the precipitate with glacial

acetic acid, keeping the solution ice-cool. The precipitate of cesium bismuth iodide is dried and weighed. (Cs) The radioactivity of cesium-137 in this fraction is estimated from the photopeak (0.66Mev, 92%) corresponding to the daughter nuclide Ba^{137m} (2.6 min.) with the gamma-ray spectrometer. However, if we estimate the activity of cesium-137 with the beta-ray low background counter, we must pay special attention to the possible contamination of potassium-40 when the activity of cesium-137 in the sample is low. The $HBiI_4$ reagent is prepared according to the method described by Evans(4) by dissoliving 10g of BiI_4 in 50ml of 55 percent HI. The presence of some free iodine would not interfere seriously with the precipitation.

12) If greater decontamination from other fission product such as rubidium is desired, we may follow the following procedure.(4)

Dissolve the above precipitate in 6 drops of conc. HCl by heating to boiling. Add rubidium carrier (10 mg/ml). Cool with ice and add $HBiI_4$ reagent. Allow to stand for several minutes and centrifuge. Wash the precipitate with glacial acetic acid, filter onto a weighed filter paper disk, and wash successively with 5ml portions of absolute ethanol and ether. Dry at 110°C for 10 min. and weigh as $Cs_3Bi_2I_9$. Dissolve this precipitate again in about 2ml of 6M HNO_3. Boil to remove I_2, add 1ml of conc. HNO_3 and 3ml of H_2O and cool the solution. To the cool solution add 10ml of ethanol (the solution should remain clear), 0.5ml of 0.5M chloroplatinic acid H_2PtCl_6 and 7ml more of alcohol. The resulating precipitate of cesium chloroplatinate is washed with alcohol, filtered onto a filter paper disk of Whatman No.50 paper, washed with ether, dried at 110°C, and weighed as Cs_2PtCl_6.

13) To the precipitate of strontium and calcium obtained in step (6) add distilled water and 90% nitric acid slowly. Keeping the concentration of nitric acid at 75%, shake or stir well for 30 min.

centrifuge at 2,000rpm for 10 min, and discard the supernatant. At this concentration strontium precipitates, but most of the calcium may remain in the solution. Repeat this procedure again at half the original volume.(2)

14) The precipitate is transferred to 100ml beaker with H_2O. Evaporate to dryness to expell nitric acid and dissolve in 50ml of distilled water.

15) To the solution, add about 10 drops of Y or Fe^{+++} carrier (10mg/ml) and heat to the incipient boiling point and make the solution basic with CO_2 free ammonium hydroxide to precipitate the hydroxides of rare earth element. Repeat the precipitation to recover strontium in the filtrate. Check the radioactivity of the precipitate obtained in this step.

16) Make the combined filtrates neutral with 6N nitric acid and add 2ml of 6M ammonium acetate and 1ml of 6M acetic acid. Add 15 drops of Ba^{++} carrier (50mg/ml) Heating the solution, add 1ml of 1.5N sodium chromate drop by drop stirring well. Continue heating the solution nearly to boiling for several minutes, allow it to cool and filter off the precipitate of barium chromate. The contamination of Ba^{140} and Ra may be eliminated.

17) Make the filtrate neutral to basic with ammonium hydroxide and add 5ml of ammonium carbonate and 95% ethanol. After stirring well, digest with heating until the precipitation of strontium carbonate is complete.

18) The precipitate of strontium carbonate is dried at 100 - 110°C for 20 minutes and weighed. (Sr) The sample is stored for about 20 days and when Sr^{90} and Y^{90} reached an equillibrium state the beta activity of the sample is measured. $[Sr^{89} + (Sr^{90} + Y^{90})]$ Then the sample is dissolved in 2ml of 6N HCl and used for Y^{90}

separation by ion exchange method.

19) After conditioning the Dowex 50 resin of about 100 - 200 mesh the ion exchange resin in H-form is filled in the glass tube of diameter about 2.5cm up to a height of about 12cm. Pass 200ml of distilled water through the column, and then convert the resin into ammonium form by passing 400ml of ammonium citrate at pH = 6.0 and wash with 400ml of distilled water at the speed of flow about 1l per hour.

20) The sample in which Sr^{90} and Y^{90} reached equillibrium is poured into the column slowly. Y^{90} is milked from Sr^{90} on the column by passing 50ml of citric acid at pH = 2.0 and 350ml of citric acid at pH = 3.8. To the combined washes about 10mg of Y-carrier is added and the solution is heated and 30ml of saturated oxalic acid is added. The solution is digested with heating at low for 10 minutes and then allowed to cool and filtered, washing with 10% $H_2C_2O_4$ solution. The precipitate of yttrium oxalate is ignited and ashed and the beta activity of this sample is measured immediately. From the decay rate of Y^{90} with half life of about 64 hours, the existence of Y^{90} is confirmed and the initial activity at the time of separation from Sr^{90} is estimated by back extrapolation. This activity of Y^{90} may be considered to correspond to that of Sr^{90}.

21) The yield in the radiochemical analysis may be estimated in the following way. Divide the original sample into two equal portions. ~~To one portion~~ To one portion (A) add a known activity of "a" μc of the radioactive nuclide to be detected, while the other portion (B) may be analysed without any addition of the radioactivity. The analysis of these two portions should be carried out almost simultaneously in parallel step by step using the same reagents. Suppose the activity of the nuclide detected after the final analysis is "c" μc in the portion (A) and "b" μc in the portion (B),

Then the final yield of this analysis may be estimated to be $\frac{c-b}{a} \times 100\%$. In the above analysis, the yield of Sr^{90} may be estimated to be about 60 - 70%, while that of Cs^{137} about 70 - 80%

However, later the above method was simplified and after the step(3) in the above the strontium was sperated from the 75% nitric acid solution according to the method described in the step (13) and the steps described thereafter was followed for the detection of Sr^{90}. The spernatant solution after the separation of strontium nitrate from the 75% nitric acid was used for the analysis of cesium. After scavenging with Fe^{+++} and Ba^{+++} carrier, cesium was precipitated as cesium bismuth iodide with $NaBiI_4$ instead of $HBiI_4$ reagent. The $NaBiI_4$ reagent was prepared by dissolving 9 grams of powered bismuth trioxide in 300ml of a saturated solution of sodium iodide in concentrated acetic acid according to the method described by Hara.(3) After heating for sometime, the soltion is rapidly filtered through a glass filter equipped with a foot and the filtrate is stored in a glass-stoppered bottle in a cold dark place and used as the $NaBiI_4$ reagent.

(II) Results of Measurements[10]

In Table 1 some of the nuclear detonation tests conduced by different countries up to the end of August 1961 are listed. In 1958 it was rather difficult to identify smaller tests because too many large tests of megaton size have been conducted by the United States as well as by the Soviet Union. Therefore only the major ones that have been detected by the shock wave records in the microbarographs of the meteorological stations in Japan are listed in the table for ~~them~~ that year.

In Table 2(a), the number of tests up to the end of 1958 reported as announced by the United States are listed and in Table 2(b) the estimated fission yield in kilotons assuming the past thermnuclear tests to have consisted on the average of 50% fission and 50% fusion is shown.[1]

(Table 1)

(Table 2)

After the cessation of nuclear tests was announced by the three major powers possessing nuclear weapons, the United States, the United Kingdom and the Soviet Union at the end of October 1958, the only tests conducted since then up to the end of August 1961 were the French ones in Sahara: three in 1960 and one in 1961. Therefore, during the past eight years period from 1954 to 1961 the only year in which no nuclear tests were conducted was 1959.

According to the New York Times,[5] somewhat different estimates are given. A total of 165 tests have been conducted by the United States, of which 122 tests were conducted in the South West U.S., 37 in Eniwetok - Bikini area in the South Pacific, one off the west coast of the United States, two in Johnson Island in the Pacific and three in the South Atlantic. The total number of British tests in reported to have been 21, of which 9 tests were conducted in

Table 1 (a)

Year	Date	Country	Test site
1945	VII:16	U.S.A	Alamagordo, New Mexico
"	VIII: 5	"	Hiroshima, Japan
"	VIII: 8	"	Nagasaki, Japan
1946	VI:30	"	Bikini Atoll
"	VII:25	"	"
1948	IV - V	"	Eniwetok Atoll
1949	VIII:	U.S.S.R	Siberia
1951	I:27,28	U.S.A	Nevada
"	II: 1, 2, 6,	"	"
"	IV, V	"	Eniwetok
"	X: 3	U.S.S.R	Siberia
"	X:22	"	"
"	X:22,28,30	U.S.A	Nevada
"	XI: 1, 5,19, 29	"	"
1952	IV: 1,15,22	"	"
"	V: 1, 7,25	"	"
"	VI: 1, 5	"	"
"	X: 3	U.K	Monte Bello Is.
"	XI: 1	U.S.A	Eniwetok
"	XI: ?	"	
1953	III:17,24,31	"	Nevada
"	IX: 6,11,16, 25	"	"
"	V: 8,19,29	"	"
"	VI: 4	"	"
"	VIII:12	U.S.S.R	Siberia
"	VIII:23	"	"
"	IX:23	"	"
"	X:15	U.K	Woomera Range, Australia

Table 1 (b)

Year	Date	Country	Test site
1954	III: 1	U.S.A	Bikini Atoll
"	III:27	"	Eniwetok
"	IV: 6	"	"
"	IV:26	"	"
"	V: 5	"	"
"	V:12		
"	IX:10,26,31	U.S.S.R	Siberia
1955	II:18,22	U.S.A	Nevada
"	III: 1, 7,12, 22,23,29	"	"
"	IV: 6, 9,15	"	"
"	V: 5,15	"	"
"	V:16	U.K	Monte Bello Is.
"	V:17	U.S.A	Pacific Ocean
"	VIII: 4-IX:24	U.S.S.R	Siberia
"	XI:	"	"
"	XI:22	"	"
1956	III:21	"	"
"	IV:	"	"
"	IV:	U.K	Monte Bello Is.
"	V: 5	U.S.A	Bikini Atoll
"	V: 7	U.K	Monte Bello Is.
"	V:21	U.S.A	Eniwetok
"	V:28	"	"
"	VI:13	"	"
"	VI:26	"	"
"	VII: 3, 9,11, 21,22	"	"
"	VIII:24	U.S.S.R	Siberia
"	VIII:30	"	"
"	IX: 2	"	"
"	IX:10	"	"
"	IX:27	U.K	Maralinga, Australia
"	X:11	"	"
"	XI:17	U.S.S.R	Siberia

Table 1 (c)

Year	Date	Country	Test site
1957	I:29	U.S.S.R	Siberia
"	III: 8	"	"
"	IV: 3	"	"
"	IV: 6	"	"
"	IV:10,12	"	"
"	IV:16	"	
"	V:15	U.K	Christmas Is.
"	V:28	U.S.A	Nevada
"	VI: 1	U.K	Christmas Is.
"	VI: 2	U.S.A	Nevada
"	VI: 5	"	"
"	VI:18	"	"
"	VI:19	U.K	Christmas Is.
"	VI:24	U.S.A	Nevada
"	VII: 5	"	"
"	VII:15	"	"
"	VII:19	"	"
"	VII:24	"	"
"	VII:25	"	"
"	VIII: 7	"	"
"	VIII:18	"	"
"	VIII:22	U.S.S.R	Siberia
"	VIII:23	U.S.A	Nevada
"	VIII:30	"	"
"	IX: 2	"	"
"	IX: 8	"	"
"	IX: 9	U.S.S.R	Siberia
"	IX:14	U.K	Maralinga
"	IX:16	U.S.A	Nevada
"	IX:24	U.S.S.R	North Pole
"	X: 6	"	"
"	XII:28	"	Siberia

Table 1 (d)

Year	Date	Country	Test site
1958	II:23,27(2)	U.S.S.R	North polar region
"	III:14(2),21	"	"
"	V:12,26	U.S.A	Eniwetok
"	VI:10	"	Bikini Atoll
"	VI:14(2),27(2), 28	"	Eniwetok
"	VII:2	"	Bikini Atoll
"	VII: 5,12,26	"	Eniwetok
"	VIII:12	U.S.S.R	North polar region
"	IX:30(2)	"	"
"	X:12,15,19,20, 22,24,25	"	"
1960	II:13	France	Sahara
"	IV: 1	"	"
"	XII:28	"	"
1961	IV:25	"	"

1945 VII - 1961 VIII Total 255.

(U.S.A 165, U.K 21, U.S.S.R 65, France 4)

Table . 2(a) The Number of Nuclear Tests.

U.S.A. (129)

July 16, 1945 - First atomic bomb test, "Trinity," Alamogordo, N.M.
August 6, 1945 - Hiroshima bomb
August 9, 1945 - Nagasaki bomb
Summer of 1946 - "Crossroads" tests (2), Bikini Atoll
Spring of 1948 - "Sandstone" tests (3), Eniwetok Proving Ground
Winter of 1951 - "Ranger" tests (5), Nevada Test Site
Spring of 1951 - "Greenhouse" tests (4), Eniwetok
Fall of 1951 - "Buster-Jangle" tests (7), Nevada
Spring of 1952 - "Tumbler-Snapper" tests (8) Nevada
Fall of 1952 - "Ivy" tests (2), Eniwetok
Spring of 1953 - "Upshot-Knothole" tests (11), Nevada
Spring of 1954 - "Castle" tests (6), Eniwetok
Spring of 1955 - "Teapot" tests (14), Nevada
Spring of 1955 - "Wigwam" test (1), Pacific Ocean
Spring -Summer of 1956 - "Redwing" tests (4), Eniwetok
Spring-Summer of 1957 - "Plumb-bob" tests (24), Nevada
Spring- Summer of 1958 - "Hardtack" series, first phase (16), Eniwetok
Fall of 1958 - "Hardtack" series, second phase (19), Nevada

U.K. (21)

October 3, 1952 - First British explosion, Monte Bello Islands
October, 1953 - Two explosions, Woomera
May-June, 1956 - Two explosions, Monte Bello Islands
September-October, 1956 - Four explosions, Marelinga
May-June, 1957 - Three explosions, Christmas Island
September-October, 1957 - Three explosions, Marelinga
November 8, 1957 - One explosion, Christmas Island
April 28, 1958 - One explosion, Christmas Island
August-September, 1958 - Four explosions

U.S.S.R. (53)

September 23, 1949 - First Soviet nuclear explosion

October, 1951 - Two known tests

August, 1953 - Series of tests, two known explosions, one thermonuclear

October 26, 1954 - "Part of a series"

August 4, 1955 - One explosion

September 24, 1955 - "Part of a series"

November, 1955 - "Part of a series," two explosions, one "in megaton range"

March-April, 1956 - "Part of a series," two explosions

August-September, 1956 - "Part of a series," four explosions

November 17, 1956 - One explosion

January 19, 1957 - "Part of a series"

March-April, 1957 - "Part of a series," six explosions

August-October, 1957 - "Part of a series," five explosions

December 28, 1957 - One explosion

February-March, 1958 - Nine explosions

September-November, 1958 - Sixteen explosions

Total: 203 up to the end of 1958.

Table 2(b)

Fission Yield in Kilotons Assuming The Past Thermonuclear Test to have Consisted on The Average of 50% Fission and 50% Fusion

	Air Bursts	Ground-Surface	Water-Surface	Total
1945-1951	190	550	20	760
1952-1954	1,000	15,000	22,000	38,000
1955-1956	5,600	1,500	6,000	13,100
1957-1958	31,000	4,400	4,600	40,000
Total	37,790	21,450	32,620	91,860

Christmas Island in the Pacific, 3 in Montebello Island and 9 in Woomera-Maralinga district in Australia. The total number of tests conducted by the Soviet Union up to the end of August 1961 is reported to have been 65 in Siberia and in Novaya Zemlya in the Arctic Ocean. The total fission yields from tests 1945 to the end of 1958 are estimated in unit of megatons TNT equivalent nuclear fission to be about 32.3 megatons (35%) water surface explosions, about 37.8 megatons (42%) air explosions and 21.5 megatons (23%) ground surface explosions. Of the total of about 92 megatons, about 70% is estimated to be due to the nuclear tests conducted by the United States and the United Kingdom and about 30% by the Soviet Union.

The radioactivity precipitated with the rain in Osaka, Japan was measured 668 times during the period April 1954 to the end of August 1961. In Table 3, monthly and annual amounts of rainfall and the number of times of the rainfalls observed at the Osaka City University are shown. As can be seen in the table, the annual amount of rainfall ranged from about 1,000 to about 1,6000 mm with the monthly average of about 80 to 110 mm. The average amount per one rainfall was estimated to be about 12 to 17 mm in Osaka district.

(Table 3)

However the frequency of small amount of rainfall is much larger than the larger ones and during the period of about seven and a half years observation the percentage of rainfalls with less than 10 mm was about 60% while that with over 60 mm was only about 3%. The number of rainy days per year was about one fourth to somewhat less than one third of a year.

The results of the daily measurements of the concentaration of radioactivity per one litre of rainwater in units of cpm/l and c/l and the precipitation of radioactivity per one square meter

Table 3

	1954		1955		1956		1957		1958		1959		1960		1961	
	Rainfall (mm)	No.	Rainfall (mm)	No.	Rainfall (mm)	No.	Rainfall (mm)	No.	Rainfall (mm)	No.	Rainfall (mm)	No.	Rainfall (mm)	No	Rainfall (mm)	No.
1			32.2	5	22.9	4	45.5	8	27.7	5	93.2	5	40.0	3	51.7	5
2			75.7	6	20.0	6	43.4	4	33.6	4	124.45	11	11.2	3	44.1	6
3			118.1	12	207.0	16	41.6	3	99.4	9	122.0	14	49.55	10	67.6	5
4	41.0	3	109.4	8	112.9	9	107.2	8	138.8	9	144.3	11	165.93	9	65.1	3
5	135.6	11	117.7	8	253.3	20	67.4	9	122.3	10	208.7	10	172.3	7	120.3	7
6	288.6	15	96.9	9	119.4	10	315.4	10	78.1	7	59.2	8	148.7	8	248.0	7
7	107.8	9	78.8	6	144.68	9	380.2	14	61.9	6	246.85	10	114.4	7	50.4	7
8	12.3	2	83.4	4	108.1	6	92.2	6	52.2	4	210.05	7	143.8	6	6.0	5
9	134.6	12	58.0	5	289.1	13	243.7	12	123.5	9	84.5	9	95.8	8		
10	29.6	7	175.4	12	145.6	9	23.4	4	163.5	9	114.1	10	66.1	6		
11	32.8	5	80.8	6	52.2	6	60.2	5	57.1	6	161.45	6	64.0	7		
12	33.3	5	28.2	6	11.6	4	23.0	3	31.3	7	61.9	3	26.2	5		
total Rainfall	815.6		1054.3		1486.78		1443.2		989.4		1630.2		1097.98		653.2	
No. of Rainfall		69		89		112		86		85		104		79		45
Avg./mon.	90.7		88.0		124.89		120.3		82.5		135.85		91.50		81.7	
Avg. per one Rainfall	11.8		11.85		13.25		16.8		11.6		15.7		13.9		14.2	

in units of cpm/m^2 and c/m^2 are shown in Fig,1 and Fig.2. Fig.1(a) and Fig.2(a) (1954).

(Fig.1(a))

(Fig.2(a))

The radioactivity of rainwater was measured 69 times in 1954. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed 7 times (during the period from May to early June), once in October and once in November, or 9 times in total which corresponded to 13.1% of the total number of measurements of that year. Higher concentration than 1,000 cpm/l (6×10^{-9} curie/l) was observed only once in the middle of May. A higher precipitation per unit arear per rainfall than 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 38 times which corresponded to 55.2%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9} curie/m^2) was observed twice. Higher precipitation than 10,000 cpm/m^2 (6×10^{-8} curie/m^2) was observed once in May following a series of American tests in the Pacific. The total amount of precipitation per unit area was about 2.29×10^{-7} c/m^2 during the period from April to the end of December and the average concentration of rainwater was estimated to be about 2.81×10^{-4} μc/l in 1954.

The annual precipitation of strontium-90 was estimated to be 1.29 mμc/m^2, while that of cesium-137 to be 2.2 mμc/m^2, with the ratio of cesium-137 to strontium-90 about 1.7. The average concentration of strontium-90 was estimated to be about 1.6 μμc/l, while that of cesium-137 about 2.7 μμc/l in 1954. The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 5.6×10^{-3}, while that of cesium-137 about 9.6×10^{-3}. The above value of Sr^{90} precipitation may be estimated to be roughly about double the total of the values estimated from the decay rate of the radioactivity of each rain in 1954, assuming the fast neutron fission of U^{238}. One of the reasons for this may be assumed to be due to the existence of the inert gas such as krypton or xenon among the parent nuclides of Sr^{90} or Cs^{137} as shown in Fig.3 and the percentage of these

(Fig. 3)

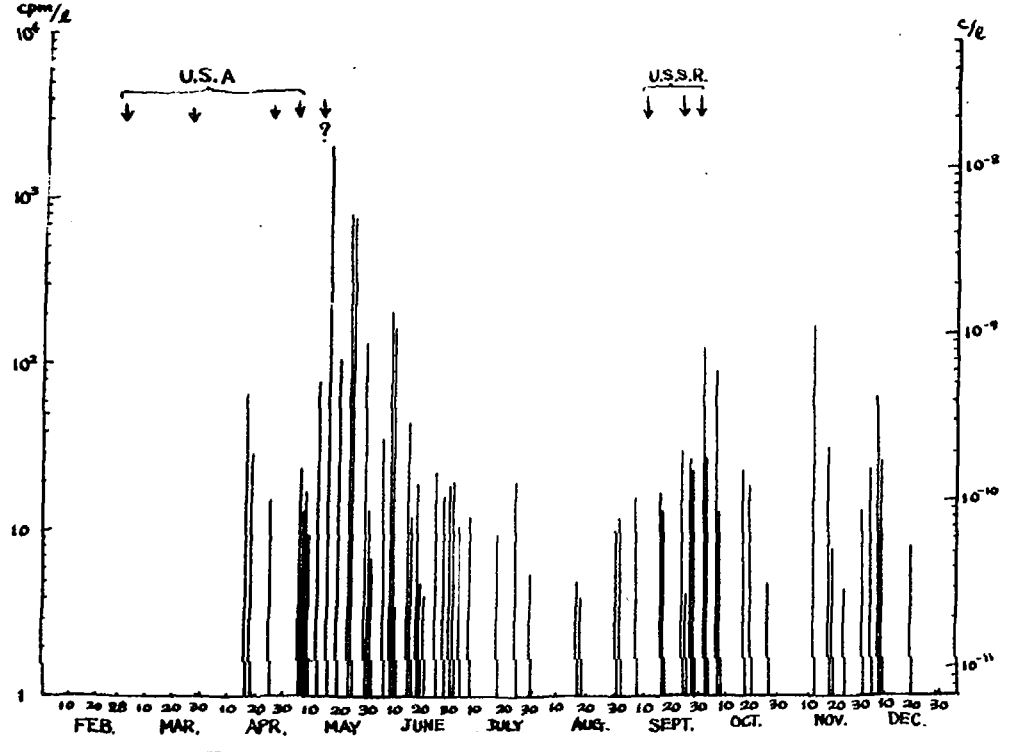


Fig 1 (a) RAIN 1954 OSAKA

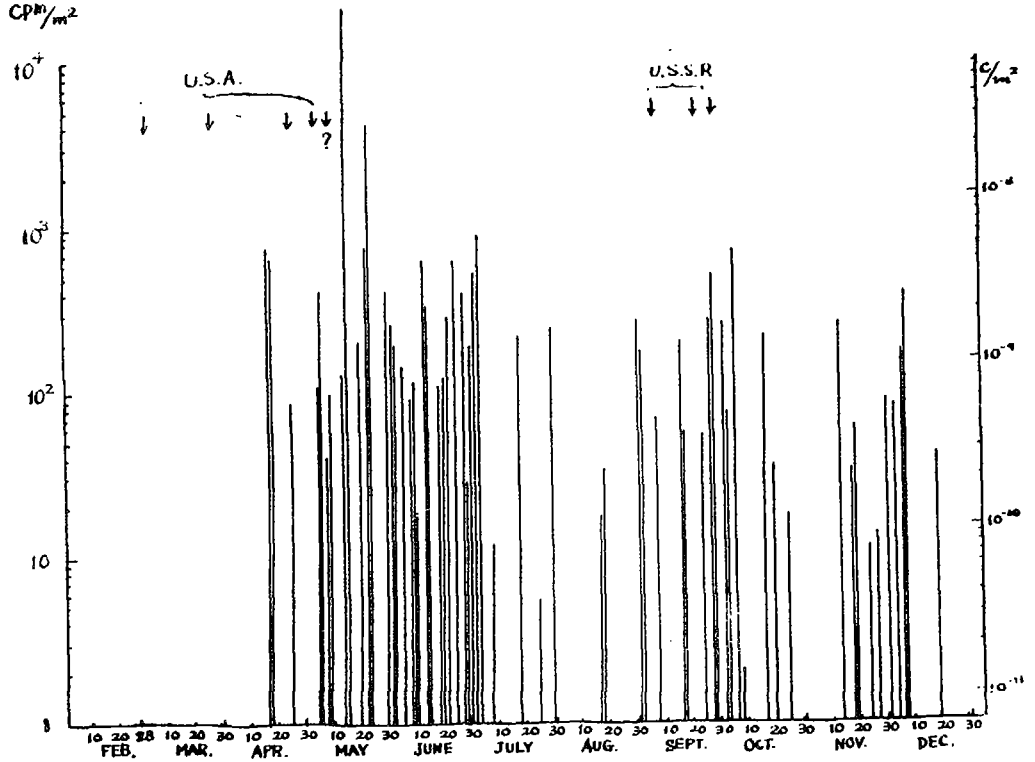


Fig. 2 (a) RAIN 1954 OSAKA

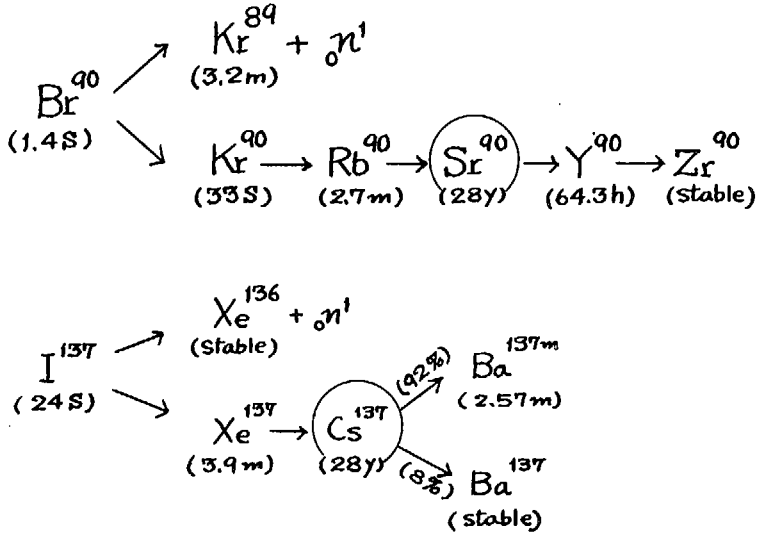


Fig. 3. Decay Chain of Sr^{90} and Cs^{137}

nuclides at remote places might be higher than that in the local fallout because of the possible higher rate of diffusion of those inert gas parents, although this degree of relative rate of diffusion may be dependent on the mode of detonation during the initial period.

Fig.1(b) and Fig.2(b) (1955).

(Fig.1(b))
(Fig.2(b))

The radioactivity of rainwater was measured 89 times in 1955. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed once in each month of January, February and April and 7 times during the period November to December, or 10 times in total which corresponded to 11.2%. Higher concentration than 1,000 cpm/l (6×10^{-9} curie/l) was observed only once towards the end of November. A higher precipitation per rainfall than 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 34 times, which corresponded to 39%. Higher precipitation than 1,000 cpm/m (6.0×10^{-9} curie/m^2) was observed 3 times.

The total amount of precipitation per unit area was about 1.64×10^{-7} c/m^2 and the average concentration of rainwater was estimated to be about 1.55×10^{-4} μc/l in 1955.

The annual precipitation of strontium-90 was estimated to be 1.69 mμc/m^2, while that cesium-137 2.88 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 1.7. The average concentration of strontium-90 was estimated to be about 1.6 μμc/l while that of cesium-137 about 2.7 μμc/l in 1955. The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 1.0×10^{-2}, while that of cesium-137 about 1.8×10^{-2}.

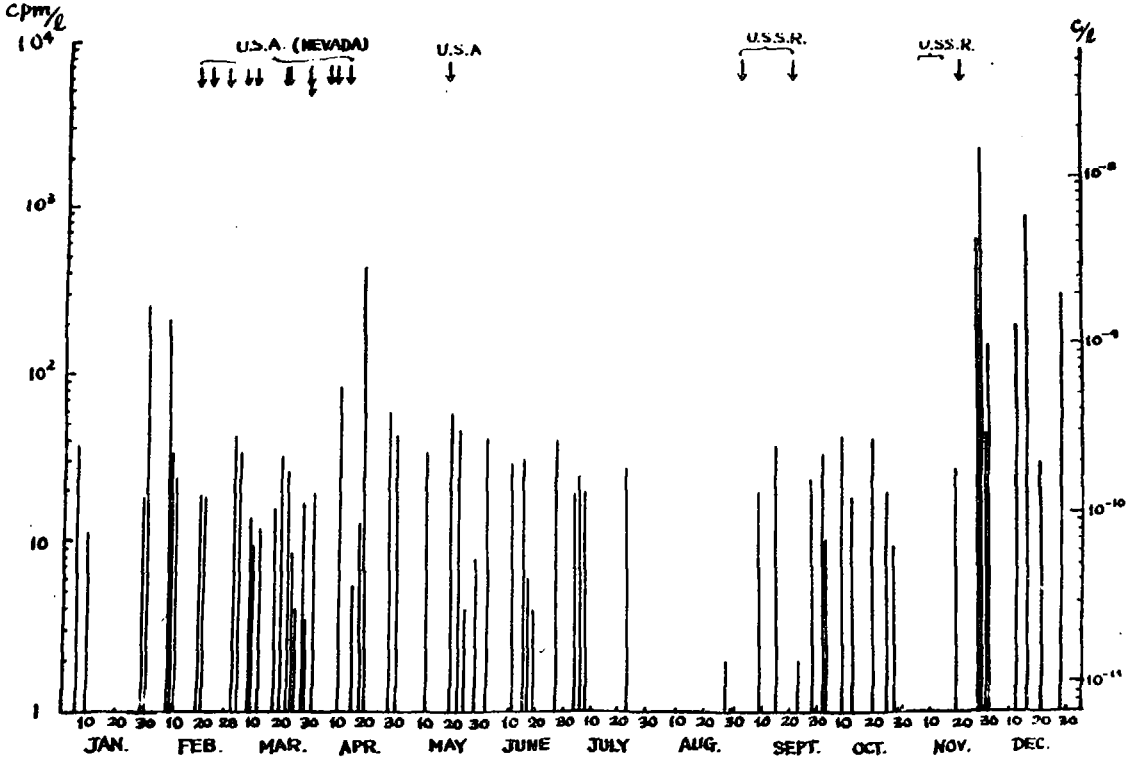


Fig. 1. (b) RAIN 1955 OSAKA

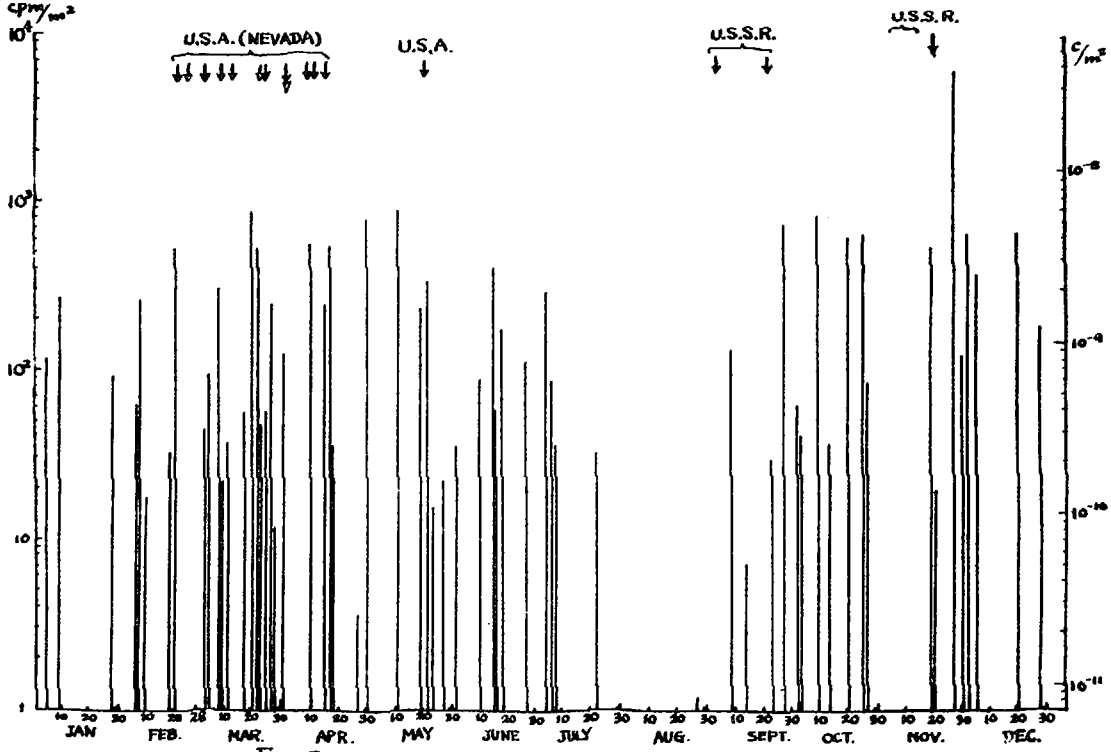


Fig. 2. (b) RAIN 1955 OSAKA

Fig.1(c) and Fig.2(c) (1956)

(Fig.1(c))

(Fig.2(c))

The radioactivity of rainwater was measured 112 times in 1956. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed once in January and in August, twice in each month of October, November and December, three times in June and in July, 4 times in March, 6 times in May, 9 times in September, or 34 times in total which corresponded to 30.4%. As compared with the previous two years, the frequency of higher radioactive rainfalls has increased. Higher concentration than 1,000 cpm/l (6×10^{-9} curie/l) was observed once in March and in June, which corresponded to 1.8%. A higher precipitation per unit area per rainfall than 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 77 times, which corresponded to 68.7%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9} curie/m^2) was observed 23 times, which corresponded to 20.5%.

The annual total amount of precipitation per unit area was 4.94×10^{-7} c/m^2 and the average concentration of rainwater was estimated to be about 3.34×10^{-4} μc/l in 1956.

The annual precipitation of strontium-90 was 2.27 mμc/m^2 while that of cesium-137 was 3.84 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 1.7. The average concentration strontium-90 was estimated to be about 1.5 μμc/l while that of cesium-137 about 2.6 μμc/l in 1956. The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 4.7×10^{-3}, while that of cesium-137 about 7.7×10^{-3}.

Fig.1(d) and Fig.2(d) (1957)

(Fig.1(d))

(Fig.2(d))

The radioactivity of rainwater was measured 86 times in 1957. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed once in each month of February, July, August and November, twice in each month of January, March, May and

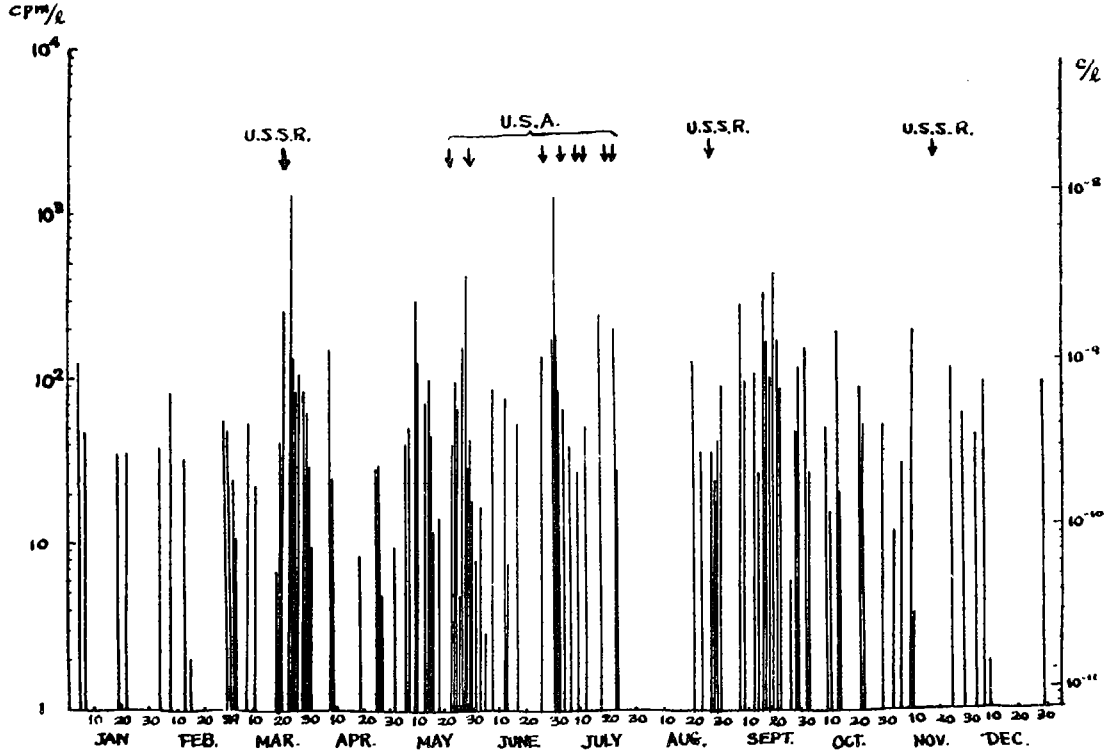


Fig. 1. (C) : RAIN 1956 OSAKA

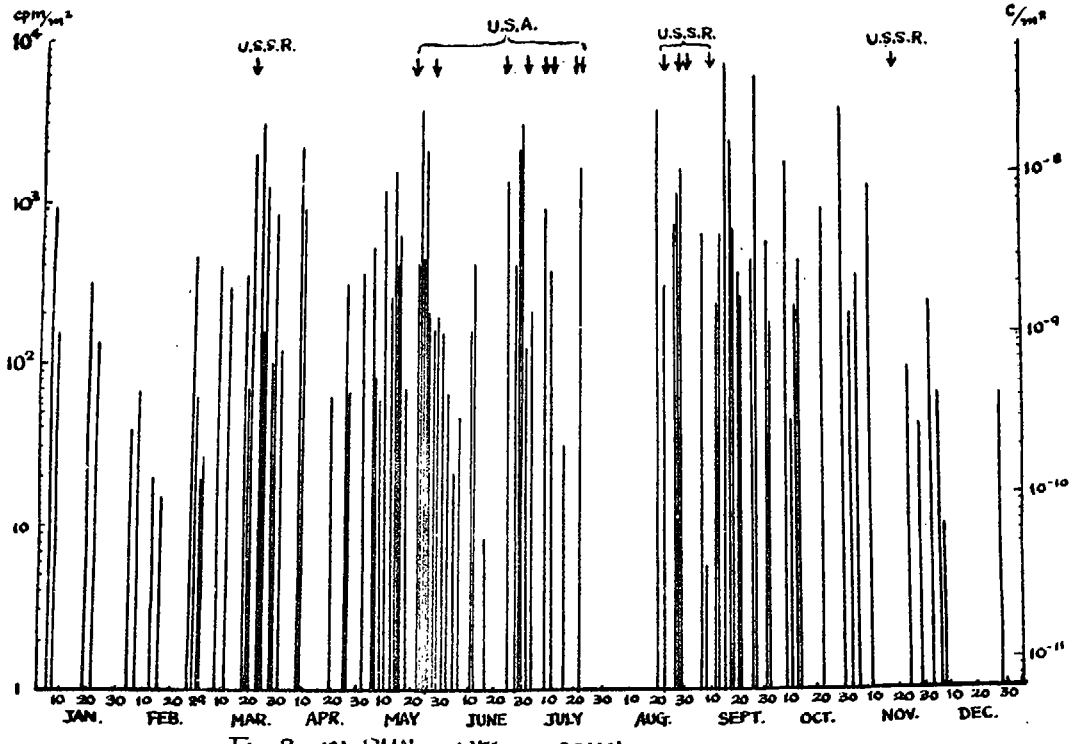


Fig. 2. (C) RAIN 1956 OSAKA

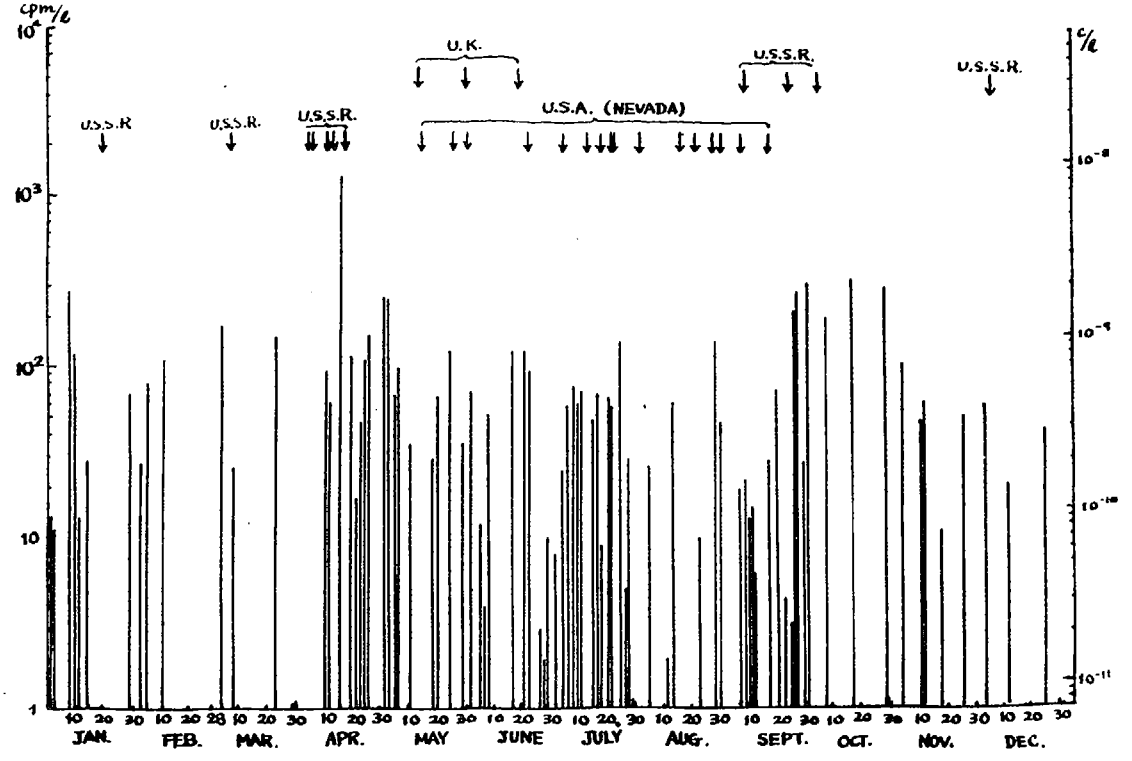


Fig. 1. (d) RAIN 1957 OSAKA

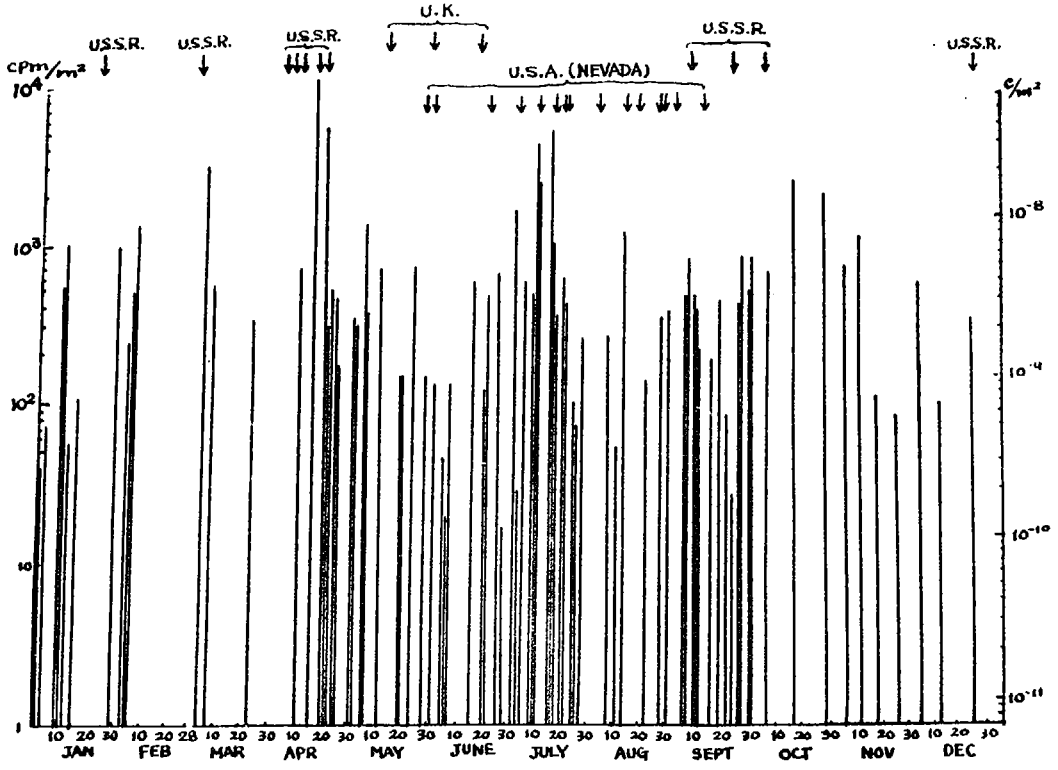


Fig. 2. (d) RAIN 1957 OSAKA

June, three times in September and in December, 5 times in April, or 23 times in total which corresponded to about 26.8%. Higher concentration than 1,000 cpm/l (6×10^{-9} curie/l) was observed only once in April. The general tendency of increase of frequency of higher radioactive rainfalls was more or less similar to that in 1956 and the frequency of rainfalls with the concentration higher than 100 cpm/l in this year was observed to be roughly about 2.5 times higher than that in 1954 or 1955.

A higher precipitation per unit area per rainfall than 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 72 times which corresponded to about 82.6%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9} curie/m^2) was observed 15 times which corresponded to about 17.6%. Higher precipitation than 10,000 cpm/m^2 (6×10^{-8} curie/m^2) was observed once in April during the period of Russian tests.

The annual total amount of precipitation per unit area was 3.67×10^{-7} c/m^2 and the average concentration of rainwater was estimated to be about 2.5×10^{-4} μc/l in 1957.

The annual precipitation of strontium-90 was 2.53 mμc/m^2 while that of cesium-137 was 4.96 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 2.0. The average concentration of strontium-90 was estimated to be about 1.7 μμc/l, while that of cesium-137 about 3.4 μμc/l in 1957.

The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 6.9×10^{-3}, while that of cesium-137 about 1.4×10^{-2}.

From the analysis of the soils about 5 to 15 cm layer from the surface which were sampled at the two different parts of the ground of Osaka City University in January 1957, about 3 mμc/m^2 and 4.25 mμc/m^2 of Sr^{90} were detected with the extraction by ammonium acetate.(2) If we assume this amount corresponds to about 70% of the total,(6) the total deposition may be estimated to be roughly about 4.3 mμc/m^2 and 6 mμc/m^2. Therefore the plausible amount of cumulative deposition of Sr^{90} in early

1957 may be estimated to be roughly in the range of about 4.3 to 6 mc/km^2 in Osaka district.

Fig.1(e) and Fig.2(e) (1958)

(Fig.1(e))

(Fig.2(e))

The radioactivity of rainwater was measured 84 times in 1958. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed 55 times out of 84 times of measurements, which corresponded to about 65.4%. In more than one half of the rainfalls, higher radioactive concentration than 100 cpm/l was measured. Higher concentration than 1,000 cpm/l was observed 10 times which corresponded to about 11.9%. In other words, higher concentrations were observed most frequently as compared with the previous years. This may be most probably due to the repeated large scale tests conducted by the United States and the Soviet Union prior to the cessation of tests at the end of October 1958. A higher precipitation per unit area per rainfall than 100 cpm/m^2 (6×10^{-10}curie/m^2) was observed 68 times which corresponded to about 81%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9}curie/m^2) was observed 33 times which was about 39.3% and higher precipitation than 10,000 cpm/m^2 (6×10^{-8}curie/m^2) was observed 9 times which was about 10.7% of the total number of measurements in 1958.

The annual total amount of precipitation per unit area was 3.01×10^{-6} c/m^2 and the average concentration of rainwater was estimated to be about 3.2×10^{-3} μc/l in 1958, which was the highest and about one order of magnitude higher as compared with the previous years. However, this may be ascribed to a relatively higher percentage of short lived fission products included in rather fresh samples.

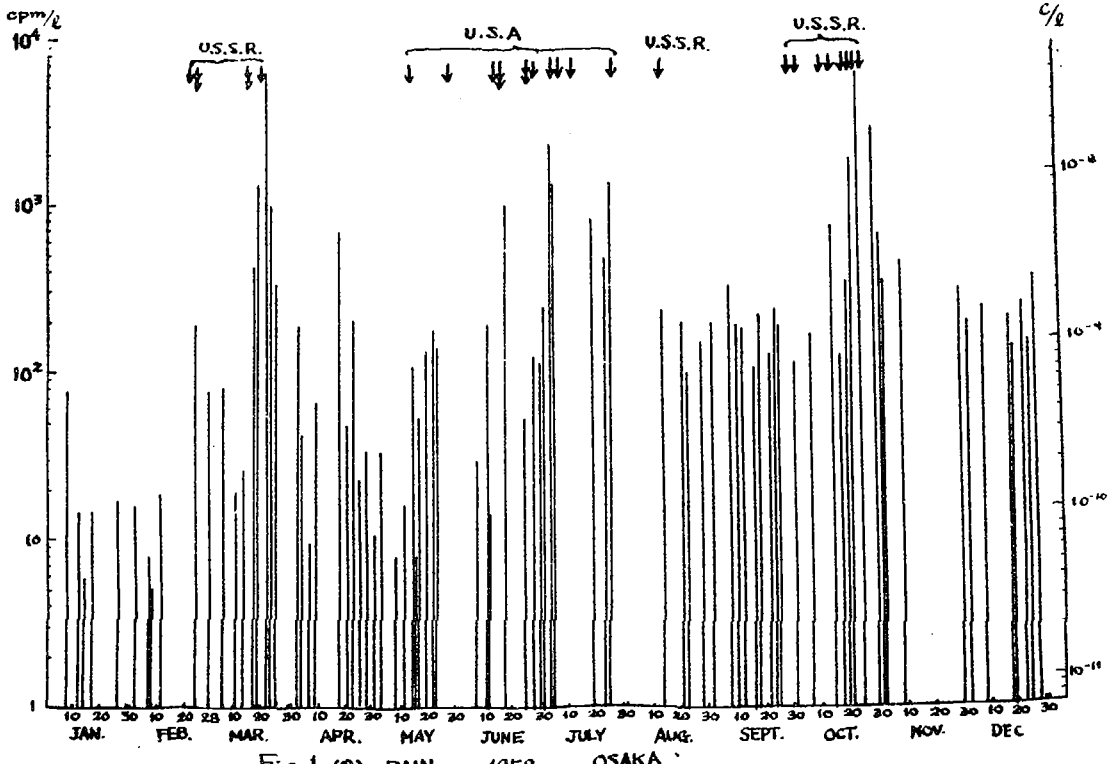


Fig. 1. (e) RAIN 1958 OSAKA

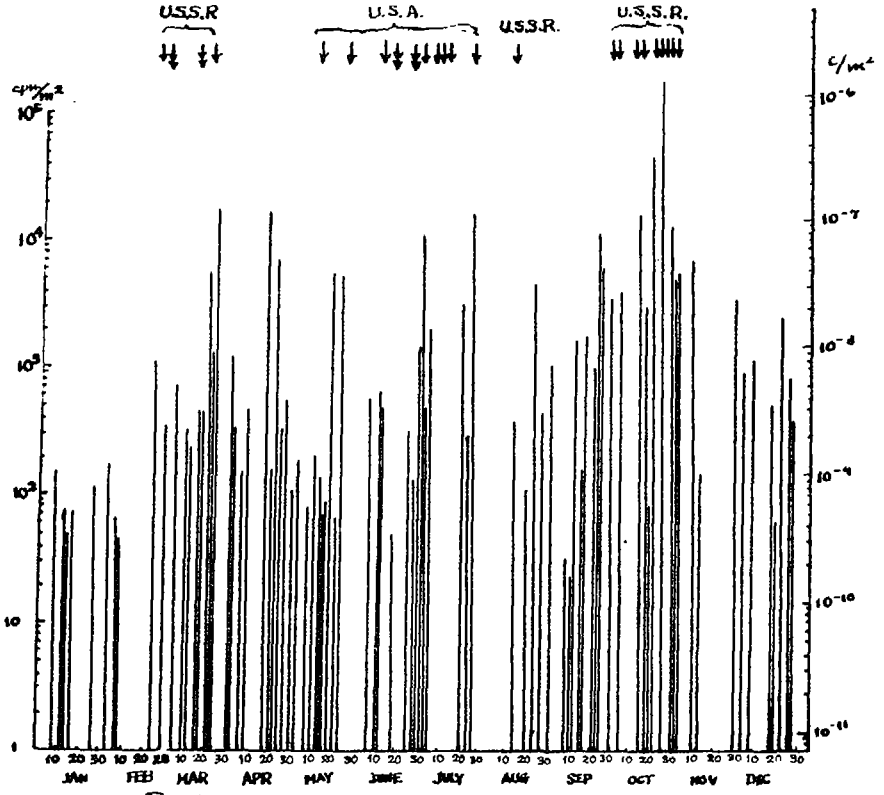


Fig. 2. (c) : RAIN 1958 OSAKA

The annual precipitation of strontium-90 was 3.08 mμc/m^2, while that of cesium-137 was 6.23 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 2.0. The average concentration of strontium-90 was estimated to be about 3.1 μμc/l while that of cesium-137 about 6.3 μμc/l in 1958.

The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 1.1 x 10^{-3} while that of cesium-137 about 2.1 x 10^{-3}.

From early spring (Feb.) to summer July 1958 the radiochemical analysis of rainwater was conducted especially to estimate the ratio of cesium-137 to strontium-90 and it was observed to vary from 1.7 to 2.5 with the average value of about 2.3 during this period.

Fig.1(f) and Fig.2(f) (1959)

(Fig.1(f))
(Fig.2(f))

The radioactivity of rainwater was measured 104 times in 1959. A higher concentration than 100 cpm/l (6 x 10^{-10} curie/l) was observed 5 times in January, 10 times in February and in March, 9 times in April, 6 times in May, 7 times in June, 8 times in July, twice in August, once in September, or 58 times in total which corresponded to about 55.7%. Higher concentration than 1,000 cpm/l (6 x 10^{-9} curie/l) was observed twice. Although no nuclear explosion tests have been conducted in this year, a high degree of radioactive contamination of rainwater almost comparable to the previous year was observed to continue until August. Usually a higher degree of strapospheric fallout may be expected at the central part of Japan during the period from winter to spring when the jet stream is at the southern part of Japan and a lower degree of fallout in summer when the jet stream is at the northern part of Japan. However, the unusally high degree of fallout

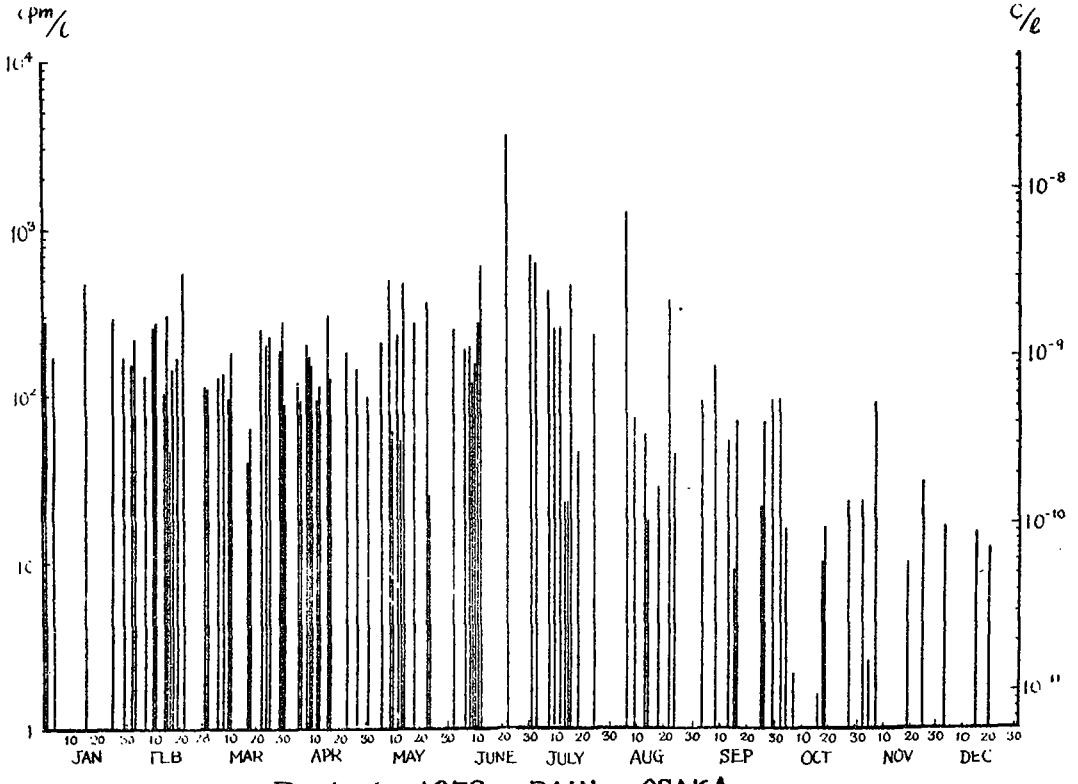


Fig. 1. (f): 1959 RAIN OSAKA

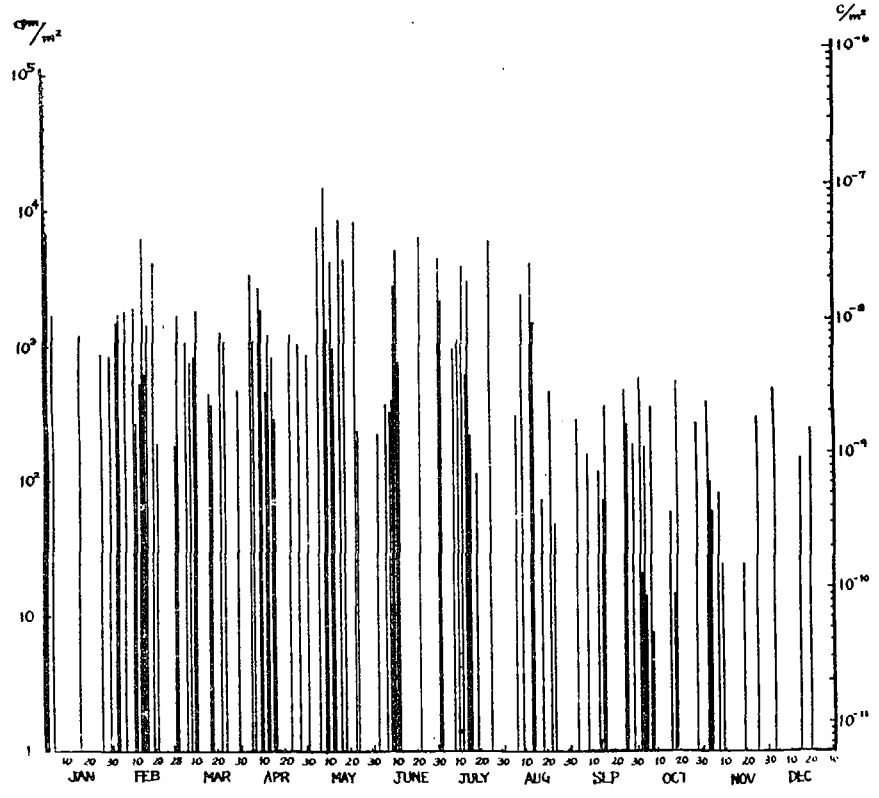


Fig. 2.(f) : 1954 RAIN OSAKA

in the first half of 1959 may be ascribed to the troposperic fallout due to the intense nuclear testings in the fall of 1958 coupled with the higher degree of stratospheric fallout usually observed in Spring.

A decreasing tendency of the radioactive contaimination of rainwater due to the fission products of relative short half lives which may be asctibed to the tropospheric fallout began to show up distinctly after August. Judging from these findings it appears more plausible to assume that the half deposition time of tropospheric fallout would be of the order of about a month or so and not very much shorter than one month. A higher precipitation per unit area per rainfall than 100 cpm/m^2 (6 x 10^{-10} curie/m^2) was observed 92 times which corresponded to about 87.5%. Higher precipitation than 1,000 cpm/m^2 (6 x 10^{-9} curie/m^2) was observed 46 times which corresponded to about 44.3%. Higher precipitation than 10,000 cpm/m^2 (6 x 10^{-8} curie/m^2) was observed once in 1959. The annual total amount of precipitation per unit area was 1.064 x 10^{-6} c/m^2 and the average radioactive concentration of rainwater was estimated to be about 6.56 x 10^{-4} μc/l in 1959.

The annual precipitation of strontium-90 was 3.34 mμc/m^2 while that of cesium-137 was 7.63 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 2.3. The average concentration of strontium-90 was estimated to be about 2.0 μμc/l while that of cesium-137 about 4.7 μμc/l in 1959. The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 3.1 x 10^{-3} while that of cesium-137 about 7.1 x 10^{-3}.

According to K.Kodaira, et al,[47] the exchangeable Sr^{90} in the agricultural soil of the Pacific side of the Japan mainland which was sampled in June 1959 was 10 mc/km^2 on the average with the highest value of 13 mc/km^2 and the lowest value of 7 mc/km^2 with the exception of one site where 26 mc/km^2 was detected, while that

of the Japan Sea side of the Japan mainland was 33 mc/km^2 on the average with the highest value of 36 mc/km^2 and the lowest value of 30 mc/km^2. Judging from these findings the total fallout at the Japan Sea side may be assumed to be roughly about three times higher than that at the Pacific side.

Fig.1(g) and Fig.2(g) (1960)

(Fig.1(g))
(Fig.2(g))

The radioactivity of rainwater was measured 79 times in 1960. A higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed 6 times in March, once in May and in September, or 8 times in total which corresponded to about 10.1%. Higher concentration than 1,000 cpm/l (6×10^{-9} curie/l) was observed twice in early March following the French test in Sahara in the middle of February. Although three French tests have been conducted in Sahara in this year, the radioactive contamination of rainwater due to the fission products of relatively short half lives was observed to be much lower except early March as compared with the previous years. A higher precipitation per unit area per rainfall than 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 31 times which corresponded to about 39.3%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9} curie/m^2) was observed 4 times and higher precipitation than 10,000 cpm/m^2 (6×10^{-8} curie/m^2) only once early March following the French test in Sahara in the middle of February. The annual total amount of precipitation per unit area was 1.9×10^{-7} c/m^2 and the average radioactive concentration of rainwater was estimated to be about 1.73×10^{-4} μc/l in 1960.

The annual precipitation of strontium-90 was 2.08 mμc/m^2 while that of cesium-137 was 4.30 mμc/m^2 with the ratio of cesium-137 to strontium-90 about 2.1. The average concentration of strontium-90 was estimated to be about 1.9 μμc/l while that of cesium-137 about

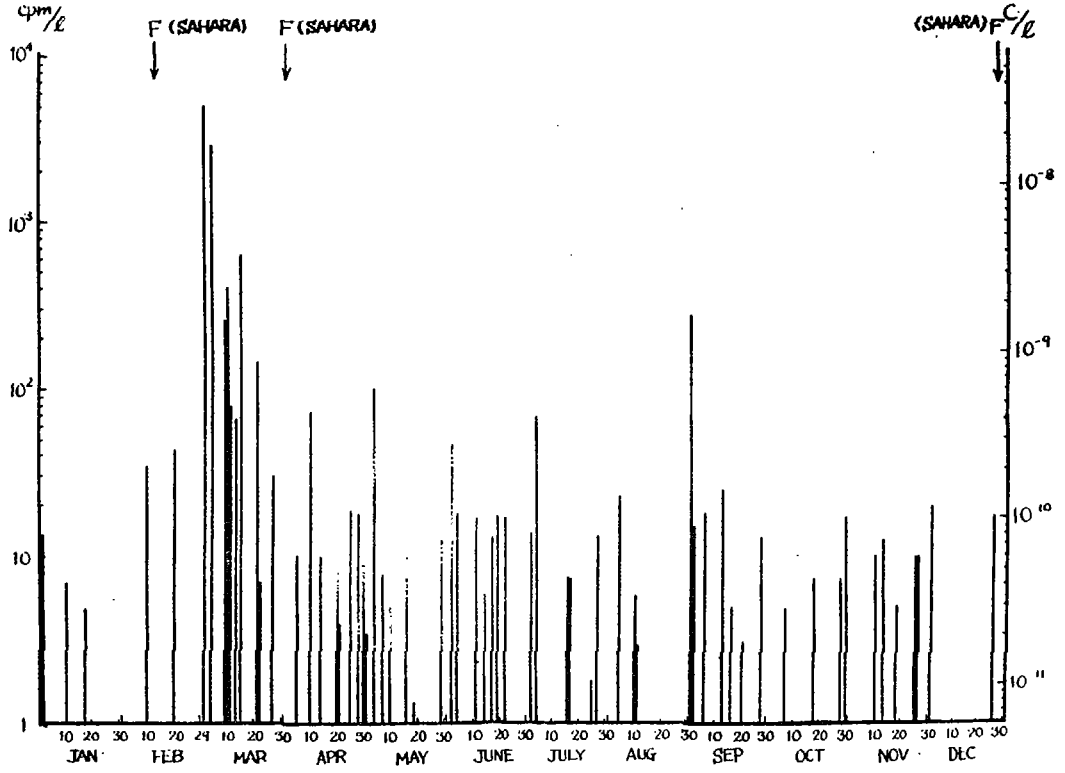


Fig. 1. (g) 1960 RAIN OSAKA

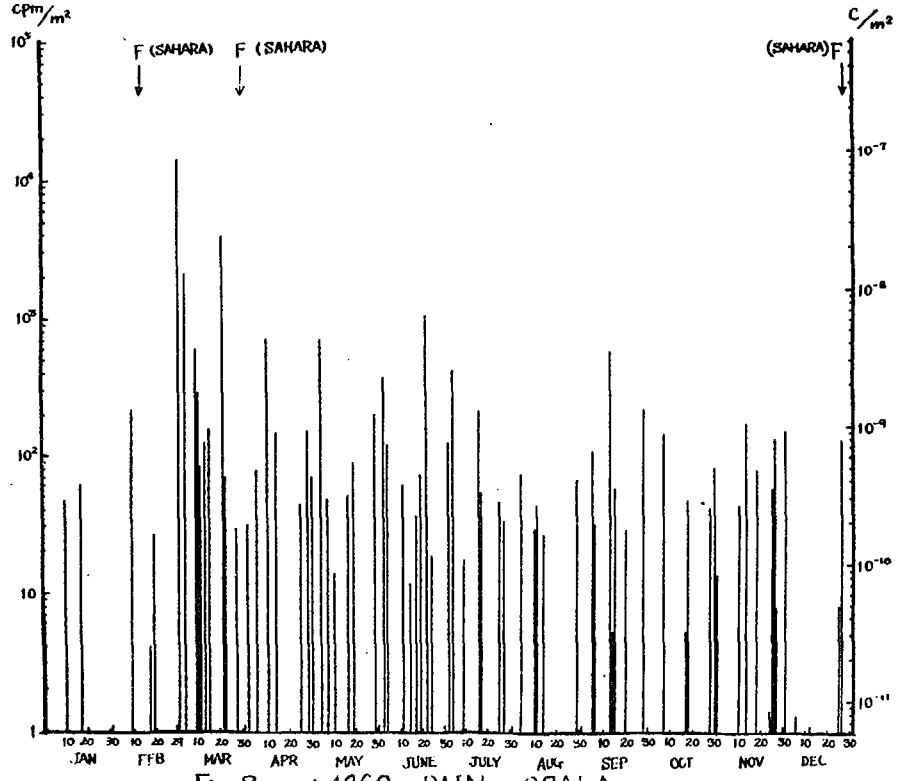


Fig. 2. (g) : 1960 RAIN OSAKA

3.9 μμc/l in 1960. The ratio of the activity of strontium-90 to the annual total of the grossactivity of each rainfall was estimated to be about 1.1×10^{-2} while that of cesium-137 about 2.3×10^{-2}.

Fig.1(h) and Fig.2(h) (1961)

(Fig.1(h))
(Fig.2(h))

The radioactivity of rainwater was measured 45 times during the period from January to the end of August 1961. The rainwater during this period was collected at Kinki University at Fuse City, Osaka Prefecture instead of at the Osaka City University, at the central part of the City.

The radioactivity was measured during this period mostly with the low background counter manufactured by the Kobe Kogyo K.K. whose background count was less than 1 cpm, but to enable the comparison of the data of this year with those of the previous years the scale of cpm in the figure was constructed so that 100 cpm correspond to 6×10^{-10} curie.

The half deposition time of longer lived fission products may be estimated to be about 400 - 800 days or roughly about 2 years from the comparison of the grossactivity of fallout during the period from Jan. to April 1960 and that during the same period in 1961 with the possible correction for relatively quickly decaying components due to French tests and taking into consideration of the decay rate of the longer lived components of the grossactivity detected in the rain.

As can be seen in the figure, a higher concentration than 100 cpm/l (6×10^{-10} curie/l) was observed only once in the middle of February and none thereafter until the end of August. Although one test was conducted by France in the Sahara desert towards the end of April, a marked decreasing tendency of the radioactive

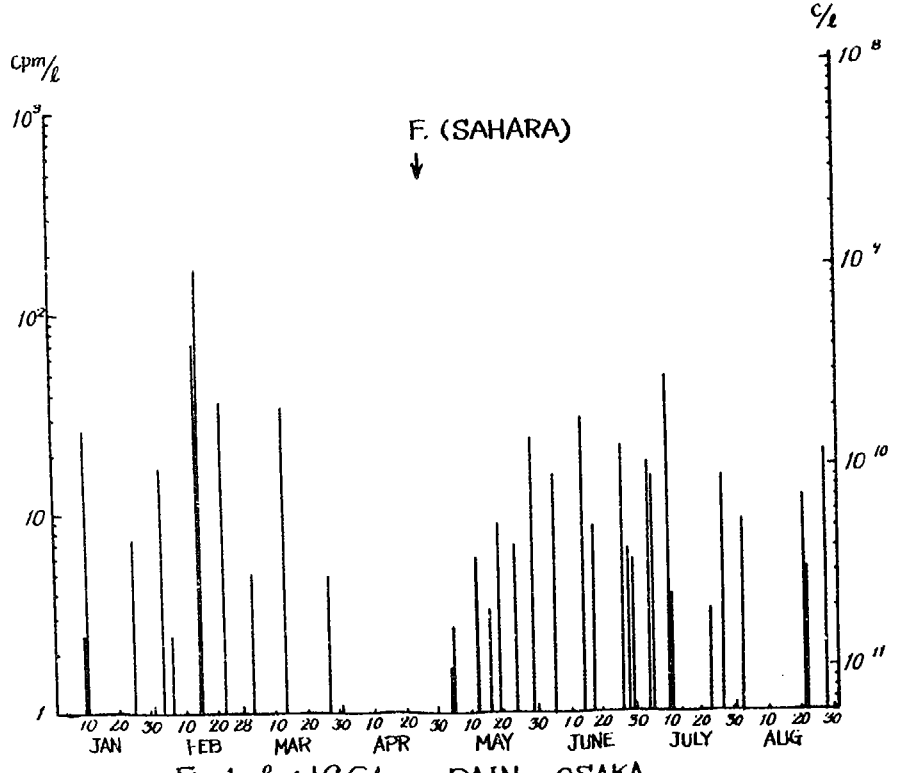


Fig. 1. (h) : 1961 RAIN OSAKA

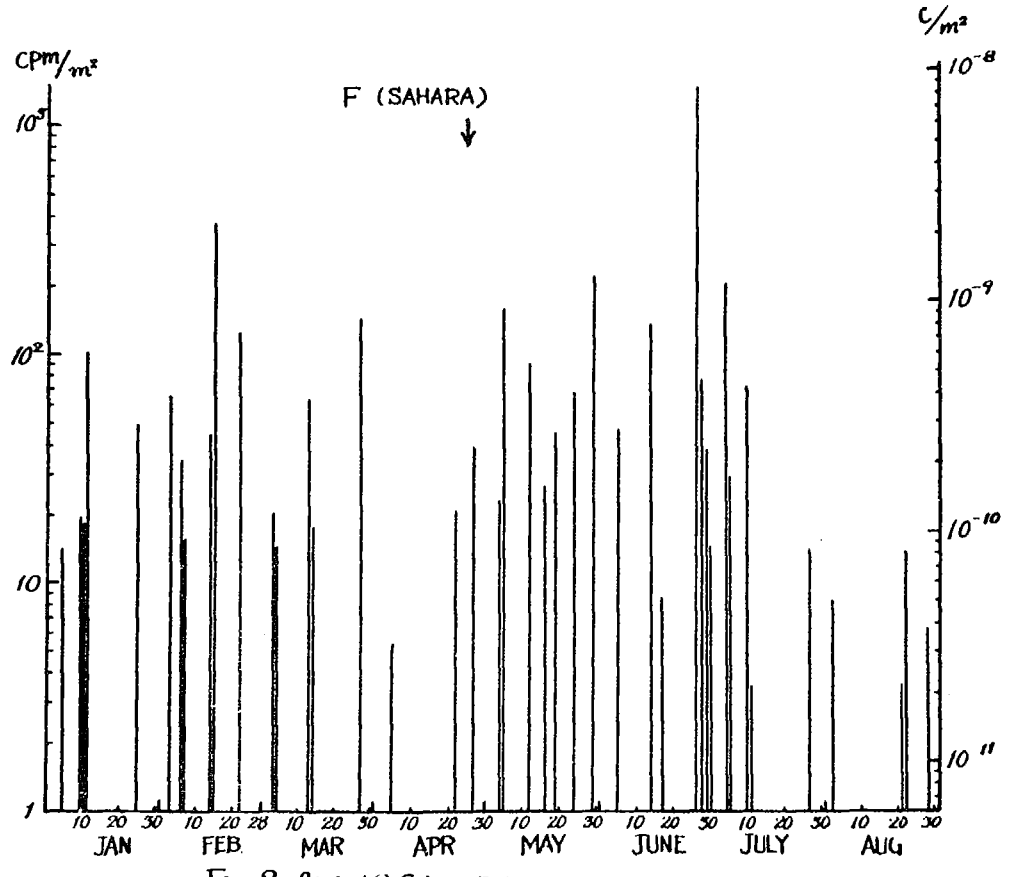


Fig.2-(h) : 1961 RAIN OSAKA

contamination of rainwater due to relatively fresh products was distinctly observed.

A higher precipitation per unit area per rainfall that 100 cpm/m^2 (6×10^{-10} curie/m^2) was observed 10 times during the period of observation in 1961 which corresponded to about 22.2%. Higher precipitation than 1,000 cpm/m^2 (6×10^{-9} curie/m^2) was observed only once in June. The total precipitation per unit area up to the end of August 1961 was 2.37×10^{-8} c/m^2 and the average radioactive concentration of rainwater was estimated to be about 3.65×10^{-5} μc/l during the period of observation.

The precipitation of strontium-90 and cesium-137 during the period from January to the end of August 1961 has not been measured. However, assuming the over all effective decay rate of the grossactivity to be about 50% from the comparison of the decay rates of grossactivities detected in 1960 and those detected during the period from January to the end of August 1961, the ratio of the activity of strontium-90 and cesium-137 to the total grossactivity during the period of observation in 1961 may be estimated to be about double higher than the ratio in 1960. From this assumption, the ratio of the activity of strontium-90 to the total grossactivity up to the end of August 1961 may be estimated to be about 2.2×10^{-2} while that of cesium-137 about 4.6×10^{-2}. Using these values, the precipitation of strontium-90 during the period from January up to the end of August 1961 may be estimated to be about 0.52 mμc/m^2 with the average concentration of about 0.8 μμc/l while that of cesium-137 about 1.1 mμc/m^2 with the average concentration of about 1.7 μμc/l.

The cumulative deposition of strontium-90 and cesium-137 estimated from the above data is shown in Fig.4, where the deposition of strontium-90 and cesium-137 is assumed to be 0.5 mc/km^2 in March 1954. As can be seen in the figure, the cumulative deposition

of Sr^{90} and Cs^{137} at the end of August 1961 may be estimated roughly at about 17.3 mc/km^2 and 33.64 mc/km^2 respectively in Osaka district, as for as judged from the results of our measurements. However, Judging from the data reported from different institutes[8] [9] in Tokyo districts, a difference by a factor of nearly two may sometimes be likely to result even in the same district and in the same month, possibly due to the difference in the sampling place and method and to the difference in chemical nature of the industrial fumes and dusts included in the rain. Therefore the above results should also be interpreted as the one that would give merely an order of magitude estimation for Osaka district. Especially the value for Cs^{137} and the ratio Cs^{137}/Sr^{90} appear to be considerably lower than those reported for Tokyo district. One of the possible causes for obtaining lower values for Cs^{137} may be considered to be the possibility that a considerable amount of Cs^{137} may be adsorbed by the particulate dust deposited in the rainwater collecting vessel during the one year period of exposure in the open and storage and that a certain fraction of Cs^{137} might be lost at the time of taking an aliquot of the rainwater from the upper part of the deep vessel as a sample for analysis.

(Fig.4)

In order to estimate the difference between the sample taken from the upper part of the vessel and that from the bottom part, an analysis of the sample taken from the bottom part after mild stirring was carried out with the method of leaching the insoluble residue with 6N hydrochloric acid. The result of the analysis indicated that the activity of Cs^{137} of the sample taken from the bottom part was higher by about 10 - 30% as compared with the results obtained with the sample from the upper part, while that

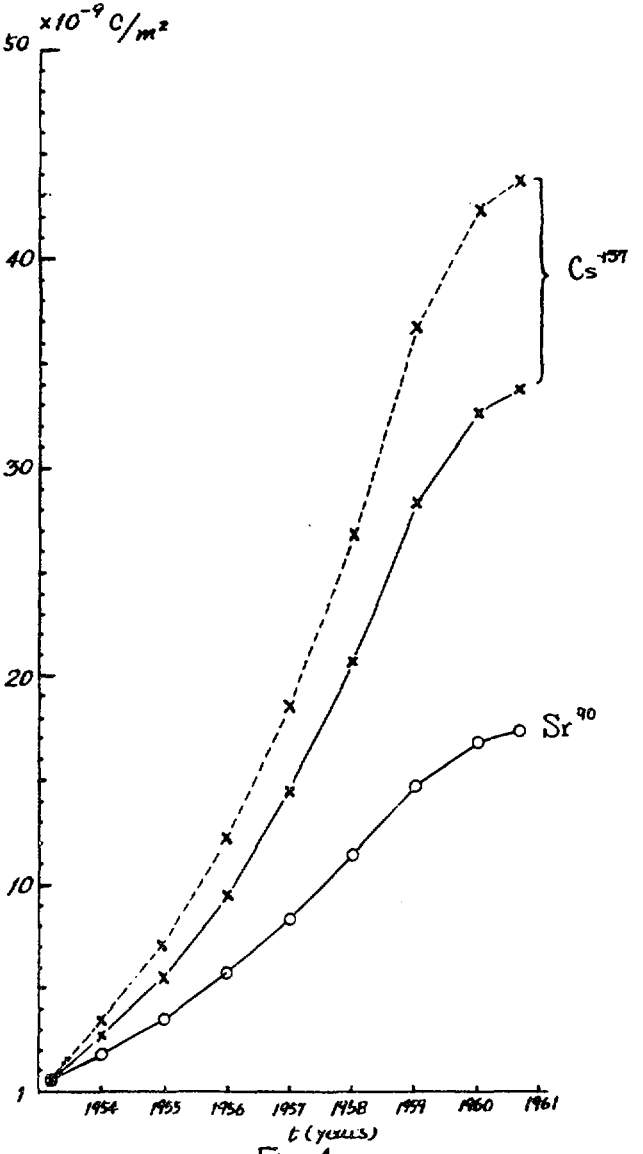

×10⁻⁹ C/m²
50
40
30
20
10
1
1954
1955
1956
1957
1958
1959
1960
1961
t (years)
Cs¹³⁷
Sr⁹⁰

Fig. 4

of Sr^{90} higher only by a few percent or less than about 10% at most. Therfore the difference in Cs^{137} may not be negligible and in order to estimate the upper limit of Cs^{137} the previously obtained results with the sample taken from the upper part of the vessel should be increased by a factor of about 30%.

(Table 4)

If we assume the upper limit of Cs^{137} to be higher than the previously obtained results by a factor of about 30%, the annual precipitation of Cs^{137} may be estimated to be about 2.8 mμc/m^2 in 1954 with the ratio Cs^{137}/Sr^{90} about 2.2, 3.75 mμc/m^2 in 1955 with the ratio Cs^{137}/Sr^{90} about 2.2, 5.0 mμc/m^2 in 1956 with ratio Cs^{137}/Sr^{90} about 2.2, 6.5 mμc/m^2 in 1957 with the ratio Cs^{137}/Sr^{90} about 2.6, 8.1 mμc/m^2 in 1958 with the ratio Cs^{137}/Sr^{90} about 2.6, 9.9 mμc/m^2 in 1959 with the ratio Cs^{137}/Sr^{90} about 3.0, 5.6 mμc/m^2 in 1960 with the ratio Cs^{137}/Sr^{90} about 2.7, 1.43 mμc/m^2 in 1961 with the ratio Cs^{137}/Sr^{90} about 2.7, and the total precipitation from April 1954 to the end of August 1961 about 43.1 mμc/m^2 with the over all ratio Cs^{137}/Sr^{90} about 2.6, and the cumulative deposition at the end of August 1961 may be estimated to be about 43.6 mμc/m^2 assuming the deposition of Cs^{137} prior to March 1954 to be about 0.5 mμc/m^2. These estimated values for Cs^{137} are also shown in the figure with the broken line as the possible upper limit for Osaka district. The results of these analysis of Cs^{137} and Sr^{90} are summarized in Table 4 together with the possible upper limit of Cs^{137} estimated using a correction factor of about 30%.

Assuming the correction factor of about 30% used to estimate the upper limit of Cs^{137}, the range of variation in the ratio of cesium-137 to strontium-90 observed from early spring to summer 1958 may be estimated to be about 1.7 - 3.3 with the average value of about 2.3 - 3.0 during the period.

(Table 5)

(Fig.5)

(Fig.6)

Table 4

	Sr^{90}			Cs^{137}			Cs^{137}/Sr^{90}
	total per unit area ($mμc/m^2$)	average concentration ($μμc/l$)	ratio to gross β	total per unit area ($mμc/m^2$)	average concentration ($μμc/l$)	ratio to gross β	
1954	1.29	1.54	5.64×10^{-3}	2.20 ~ 2.86	2.70 ~ 3.51	$9.60\times10^{-3} \sim 1.25\times10^{-2}$	1.70 ~ 2.22
1955	1.69	1.60	1.03×10^{-2}	2.88 ~ 3.77	2.73 ~ 3.55	$1.76\times10^{-2} \sim 2.29\times10^{-2}$	1.70 ~ 2.23
1956	2.27	1.53	4.60×10^{-3}	3.84 ~ 4.99	2.58 ~ 3.36	$7.78\times10^{-3} \sim 1.01\times10^{-2}$	1.69 ~ 2.20
1957	2.53	1.75	6.90×10^{-3}	4.46 ~ 6.45	3.44 ~ 4.47	$1.35\times10^{-2} \sim 1.75\times10^{-2}$	1.96 ~ 2.55
1958	3.08	3.12	1.02×10^{-3}	6.23 ~ 8.21	6.30 ~ 8.19	$2.07\times10^{-3} \sim 2.69\times10^{-3}$	2.01 ~ 2.68
1959	3.34	2.05	3.14×10^{-3}	7.63 ~ 9.92	4.68 ~ 5.96	$7.16\times10^{-3} \sim 9.31\times10^{-3}$	2.28 ~ 2.46
1960	2.08	1.89	1.09×10^{-2}	4.30 ~ 5.59	3.92 ~ 5.10	$2.26\times10^{-2} \sim 2.94\times10^{-2}$	2.06 ~ 2.64
1961 (I~III)	0.52	0.8	2.2×10^{-2}	1.1 ~ 1.43	1.7 ~ 2.21	$4.6\times10^{-2} \sim 5.98\times10^{-2}$	2.1 ~ 2.75

Table 5: Total grossactivity precipitated with rain (c/m^2)

Year / Month	1954	1955	1956	1957	1958	1959	1960	1961
1		2.9×10^{-9}	9.2×10^{-9}	1.69×10^{-8}	2.9×10^{-9}	1.17×10^{-7}	2.3×10^{-9}	1.23×10^{-9}
2		5.5×10^{-9}	4.0×10^{-9}	1.25×10^{-8}	8.5×10^{-9}	1.26×10^{-7}	1.53×10^{-9}	3.9×10^{-9}
3		1.36×10^{-8}	5.15×10^{-8}	2.45×10^{-8}	1.72×10^{-7}	8.3×10^{-8}	1.31×10^{-7}	1.56×10^{-9}
4	9.0×10^{-10}	8.7×10^{-9}	2.19×10^{-8}	1.17×10^{-7}	1.80×10^{-7}	9.6×10^{-8}	1.07×10^{-8}	3.92×10^{-10}
5	1.69×10^{-7}	1.34×10^{-8}	7.77×10^{-8}	2.61×10^{-8}	2.0×10^{-8}	3.12×10^{-7}	7.1×10^{-9}	3.80×10^{-9}
6	2.03×10^{-8}	5.2×10^{-9}	2.80×10^{-8}	1.43×10^{-8}	2.23×10^{-8}	1.0×10^{-7}	1.06×10^{-8}	1.07×10^{-8}
7	1.32×10^{-8}	7.0×10^{-10}	5.33×10^{-8}	5.17×10^{-8}	2.03×10^{-7}	1.40×10^{-7}	5.6×10^{-9}	1.92×10^{-9}
8	3.0×10^{-10}	5.1×10^{-10}	4.49×10^{-8}	6.0×10^{-9}	3.57×10^{-8}	5.48×10^{-8}	1.63×10^{-9}	1.97×10^{-10}
9	1.02×10^{-8}	5.9×10^{-9}	1.45×10^{-7}	3.62×10^{-8}	1.63×10^{-7}	1.20×10^{-8}	1.18×10^{-8}	
10	8.2×10^{-9}	1.36×10^{-8}	4.82×10^{-8}	3.58×10^{-8}	2.0×10^{-6}	1.28×10^{-8}	2.03×10^{-9}	
11	2.4×10^{-9}	8.08×10^{-8}	1.22×10^{-8}	1.91×10^{-8}	1.56×10^{-7}	5.92×10^{-9}	3.01×10^{-9}	
12	4.7×10^{-9}	1.2×10^{-8}	2.4×10^{-9}	6.6×10^{-9}	4.51×10^{-8}	5.4×10^{-9}	1.79×10^{-9}	
Total	2.29×10^{-7}	1.64×10^{-7}	4.94×10^{-7}	3.67×10^{-7}	3.01×10^{-6}	1.06×10^{-6}	1.90×10^{-7}	2.37×10^{-8}
Avg/mon.	2.54×10^{-8}	1.36×10^{-8}	4.14×10^{-8}	3.06×10^{-8}	2.51×10^{-7}	8.90×10^{-8}	1.58×10^{-8}	2.96×10^{-7}

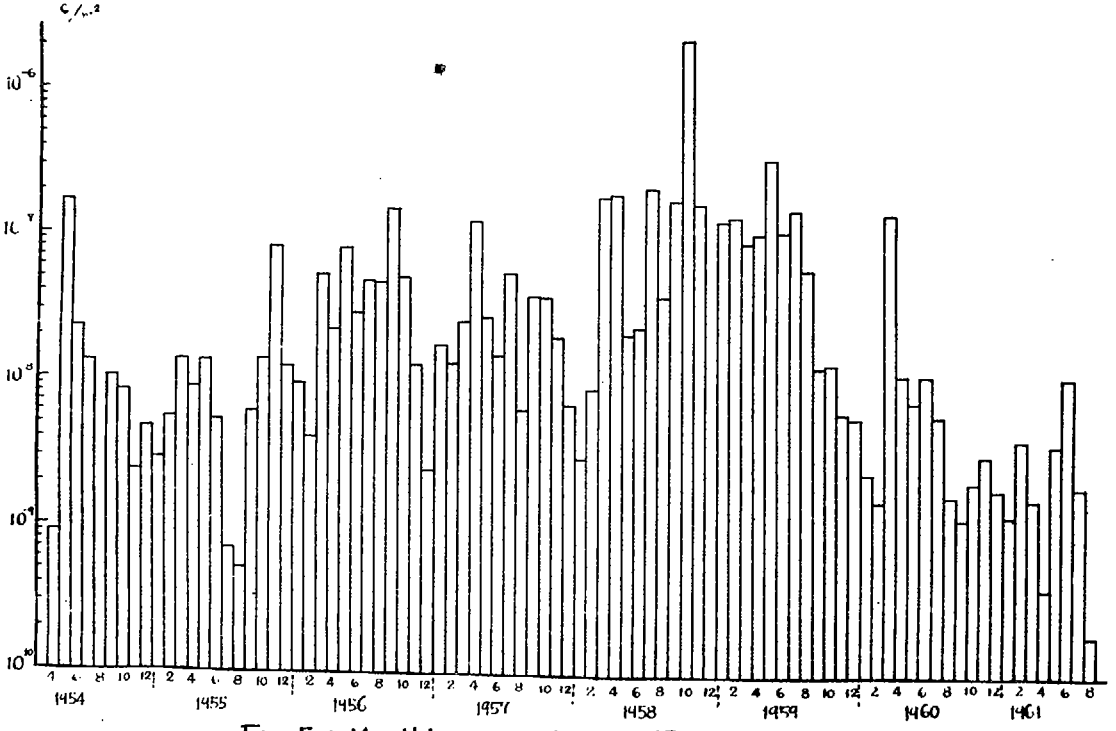
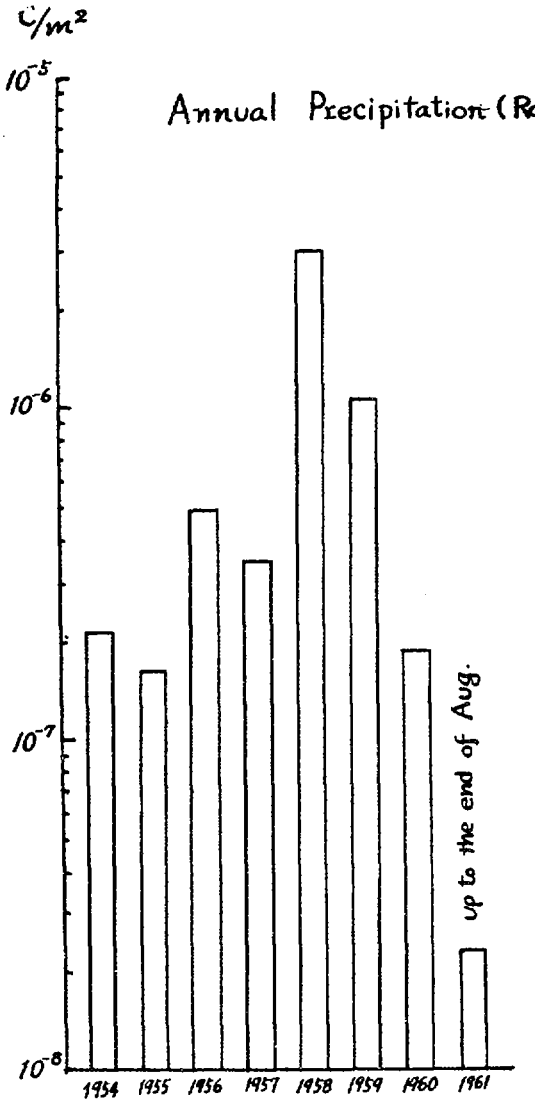


Fig. 5 : Monthly precipitation (Rain)


C/m²
10⁻⁵
10⁻⁶
10⁻⁷
10⁻⁸
Annual Precipitation (Rain)
up to the end of Aug.
1954
1955
1956
1957
1958
1959
1960
1961
Fig. 6

Monthly and annual totals of the grossactivity of rainfalls are shown in Table 5 and in Figs.5 and 6. Monthly and annual averages of the radioactive concentrations of rainwater are given in Table 6. The average radioactive concentrations were estimated by dividing the total precipitation of grossactivity per unit area by the total amount of rainfalls per unit area.

(Table 6)

The frequency distribution of different degrees of radioactives contaminations in each year is shown in Table 7 with the radioactive concentrations of rainwater and in Table 8 with the precipitations of grossactivity per unit area. The relation between the amount of rainfall and the degree of radioactive contamination of rainwater is shown in Table 9. As can be seen in the table clearly, there is a general tendency that the percentage of higher radioactive concentration of rainwater is higher with the smaller amount of rainfall, and that the percentage of lower concentration is higher with the larger amount of rainfall.

(Table 7)

(Table 8)

(Table 9)

From the above data, the grand total of the grossactivity of each rainfall per unit area during the period from April 1954 to the end of August 1961 may be estimated to be about 5.54 $\mu c/m^2$ or 5.54 c/km^2. Although there may be places where higher or lower activity might have been measured even in the same district at different parts of Japan, multiplying the above value by the total area of Japan about 3.6×10^5 km^2, the total activity carried over to Japan during the period of observation from April 1954 up to the end of August 1961 may be estimated to be roughly about two million curies or possibly three million curies because there are places where higher rate of fallout than in Osaka has been reported.

Table 6: Monthly averages of the radioactive concentration of rainwater ($\mu c/\ell$)

	1954	1955	1956	1957	1958	1959	1960	1961
1		9.0×10^{-5}	4.02×10^{-4}	3.72×10^{-4}	1.05×10^{-4}	1.26×10^{-3}	5.75×10^{-5}	2.38×10^{-5}
2		7.3×10^{-5}	2.00×10^{-4}	2.88×10^{-4}	2.53×10^{-4}	1.01×10^{-3}	1.37×10^{-4}	8.85×10^{-5}
3		1.15×10^{-4}	2.54×10^{-4}	2.79×10^{-4}	1.73×10^{-3}	8.70×10^{-4}	2.64×10^{-3}	2.30×10^{-5}
4	2.2×10^{-5}	8.0×10^{-5}	1.94×10^{-4}	1.09×10^{-3}	1.30×10^{-3}	6.65×10^{-4}	6.45×10^{-5}	6.02×10^{-6}
5	1.79×10^{-4}	1.14×10^{-4}	3.07×10^{-4}	3.98×10^{-4}	1.63×10^{-4}	1.50×10^{-3}	4.10×10^{-5}	3.16×10^{-5}
6	7.03×10^{-4}	5.4×10^{-5}	2.34×10^{-4}	4.5×10^{-5}	2.89×10^{-4}	1.69×10^{-3}	7.15×10^{-5}	4.30×10^{-6}
7	1.23×10^{-4}	9.0×10^{-6}	3.69×10^{-4}	1.36×10^{-4}	3.28×10^{-5}	5.67×10^{-4}	4.90×10^{-5}	3.80×10^{-5}
8	2.40×10^{-5}	6.05×10^{-7}	4.15×10^{-4}	6.5×10^{-5}	6.85×10^{-4}	2.61×10^{-4}	1.13×10^{-5}	3.28×10^{-5}
9	7.60×10^{-5}	1.02×10^{-4}	5.01×10^{-4}	1.49×10^{-4}	1.32×10^{-3}	1.43×10^{-4}	1.23×10^{-4}	
10	2.77×10^{-4}	7.8×10^{-5}	3.31×10^{-4}	1.53×10^{-3}	1.22×10^{-2}	1.12×10^{-4}	3.30×10^{-5}	
11	7.30×10^{-5}	7.92×10^{-4}	2.35×10^{-4}	3.18×10^{-4}	2.77×10^{-3}	3.68×10^{-5}	4.70×10^{-5}	
12	1.41×10^{-4}	4.26×10^{-4}	2.07×10^{-4}	2.87×10^{-4}	1.44×10^{-3}	8.73×10^{-5}	7.60×10^{-5}	
Av.*	2.81×10^{-4}	1.55×10^{-4}	3.34×10^{-4}	2.54×10^{-4}	3.20×10^{-3}	6.55×10^{-4}	1.73×10^{-4}	3.64×10^{-5}

* Averge concentration was estimated by dividing the total precipitation of radioactivity per unit area by the total amount of rainfall per unit area.

Table 7 : Rain

Year \ C/ℓ, cpm/ℓ, No. and %	$< 6\times10^{-11}$		6×10^{-11} ~ 3×10^{-10}		3×10^{-10} ~ 6×10^{-10}		6×10^{-10} ~ 6×10^{-9}		$6\times10^{-9} <$		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 <			
	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
1954	24	34.8	32	46.4	4	5.8	8	11.6	1		69	100
1955	36	40.5	40	45.0	3	3.37	9	10.1	1	1.12	89	〃
1956	15	13.4	37	33.0	26	23.2	32	28.6	2	1.77	112	〃
1957	12	14.0	29	33.7	22	25.6	22	25.6	1	1.16	86	〃
1958	7	8.34	15	17.9	7	8.34	45	53.5	10	11.9	84	〃
1959	7	6.73	22	21.2	17	16.4	56	53.8	2	1.92	104	〃
1960	37	46.8	29	36.7	5	6.33	6	7.6	2	2.53	79	〃
1961	30	66.6	13	28.9	1	2.22	1	2.22	0	—	45	〃

Table 8 : Rain

Year \ C/m² / cpm/m² / No. and %	< 6×10^{-11}		6×10^{-11} ~ 3×10^{-10}		3×10^{-10} ~ 6×10^{-10}		6×10^{-10} ~ 6×10^{-9}		6×10^{-9} ~ 6×10^{-8}		6×10^{-8} <		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 ~ 10000		10000 <			
	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
1954	9	13.0	13	18.8	9	13.0	36	52.0	1	1.45	1	1.45	69	100
1955	27	30.3	18	20.2	10	11.2	31	34.9	3	3.37	0	—	89	"
1956	7	6.25	14	12.5	14	12.5	54	48.2	23	20.5	0	—	112	"
1957	2	2.33	6	6.98	6	6.98	57	66.2	14	16.3	1	1.26	86	"
1958	0	—	5	5.95	11	13.1	35	41.7	24	28.6	9	10.7	84	"
1959	3	2.88	2	1.92	7	6.72	46	44.2	45	43.3	1	0.96	104	"
1960	8	10.1	22	27.8	18	22.8	27	34.2	3	3.80	1	1.26	79	"
1961	9	20.0	19	42.2	7	15.6	4	20.0	1	2.22	0		45	"

Table 9 (a) The relation between the radioactive contamination and the amount of rainfall during the period from April 1954 to December 1958.

Rainfall (m.m) \ c/ℓ, cpm/ℓ, No. and %, Rainfall	$< 6\times10^{-11}$		$6\times10^{-11} \sim 3\times10^{-10}$		$3\times10^{-10} \sim 6\times10^{-10}$		$6\times10^{-10} \sim 6\times10^{-9}$		$6\times10^{-9} <$		Total	
	0 ~ 10		10 ~ 50		50 - 100		100 ~ 1000		1000 <			
	No.	%	No.	%	No.	%	No.	%	No	%	No.	%
0 ~ 1	5	5.26	8	5.2	6	9.68	25	21.55	4	26.7	48	10.85
1 ~ 10	36	36.9	74	48.0	34	54.9	64	55.2	6	40.0	214	48.4
10 ~ 20	19	20.0	31	20.1	15	24.2	10	8.63	3	20.0	78	17.65
20 ~ 30	10	10.52	24	15.6	2	3.23	10	8.63	2	13.3	48	10.85
30 ~ 40	7	7.37	10	6.5	2	3.23	3	2.59	0	—	22	4.98
40 ~ 60	12	12.63	3	1.95	0	—	4	3.45	0	—	19	4.3
60 <	6	6.32	4	2.6	3	4.84	0	—	0	—	13	2.94
Total	95	100.0	154	100.0	62	100.0	116	100.0	15	100.0	442	100.0

Table 9(b): The relation between the radioactive contamination and the amount of rainfall during the period from January 1959 to August 1961.

Rainfall (mm) \ C/ℓ, cpm/ℓ, No. and %	$< 6 \times 10^{-11}$		$6 \times 10^{-11} \sim 3 \times 10^{-10}$		$3 \times 10^{-10} \sim 6 \times 10^{-10}$		$6 \times 10^{-10} \sim 6 \times 10^{-9}$		$6 \times 10^{-9} <$		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 <			
	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
0 ~ 1	5	6.95	6	9.24	1	4.55	4	6.35	2	50.0	18	9.8
1 ~ 10	32	44.5	35	53.9	13	59.0	34	54.0	2	50.0	116	49.4
10 ~ 20	17	23.6	12	18.5	5	22.7	14	22.2	0	—	48	18.9
20 ~ 30	6	8.33	6	9.24	2	9.1	9	14.3	0	—	23	10.6
30 ~ 40	5	6.95	1	1.54	1	4.55	1	1.59	0	—	8	4.44
40 ~ 60	3	4.16	0	—	0	—	1	1.59	0	—	4	3.44
60 <	4	5.55	5	7.7	0	—	0	—	0	—	9	3.29
Total	72	100.0	65	100.0	22	100.0	63	100.0	4	100.0	226	100.0

Table 9 (c): The relation between the radioactive contamination and the amount of rainfall during the period from April 1954 to August 1961.

Rainfall (mm) \ C/ℓ, cpm/ℓ, No. and %	$< 6\times10^{-11}$		$6\times10^{-11} \sim 3\times10^{-10}$		$3\times10^{-10} \sim 6\times10^{-10}$		$6\times10^{-10} \sim 6\times10^{-9}$		$6\times10^{-9} <$		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 <			
	No	%	No.	%	No.	%	No.	%	No.	%	No.	%
0 ~ 1	10	5.99	14	6.4	7	8.34	29	16.2	6	31.6	66	9.8
1 ~ 10	68	40.6	109	49.8	47	56.0	98	54.8	8	42.1	330	49.4
10 ~ 20	36	21.55	43	19.3	20	23.8	24	13.4	3	15.8	126	18.9
20 ~ 30	16	9.56	30	13.7	4	4.76	19	10.6	2	10.5	71	10.6
30 ~ 40	12	7.19	11	5.02	3	3.57	4	2.24	0	—	30	4.49
40 ~ 60	15	8.97	3	1.37	0	—	5	2.80	0	—	23	3.44
60 <	10	5.99	9	4.11	3	3.57	0	—	0	—	22	3.29
Total	167	100.0	219	100.0	84	100.0	179	100.0	19	100.0	668	100.0

The grand total of the activity of Sr^{90} during the period of observation may be estimated to be about 16.8 mc/km^2 while that of Cs^{137} about 33.14 - 43.1 mc/km^2 in Osaka district. Multiplying the above value per unit area by the total area of Japan, the total activity of Sr^{90} and Cs^{137} carried over to Japan may be estimated to be roughly about 6×10^3 curies of Sr^{90} and 1.2×10^4 - 1.6×10^4 curies of Cs^{137}. However, since about three times higher precipitation than the above value are estimated at the Japan Sea Side of the Japan Mainland for Sr^{90} and Cs^{137}, if we assume about double the above value as the representative figure for Japan, the total activity of Sr^{90} and Cs^{137} carried over to Japan during the period from April 1954 to the end of August 1961 may be estimated roughly at about 1.2×10^4 curies of Sr^{90} and 2.4×10^4 - 3.1×10^4 curies of Cs^{137}.

The radioactive fallout was also measured by exposing the greased paper with the surface area of about 0.1 m^2 during the period from Feb. 16, 1955 to Dec. 31, 1957. After exposing the greased paper for a certain period of time, the paper was wrapped up and completely ashed in the electric furnace. The beta activity of the ash was measured usually one to two days after sampling. The results of measurements are shown in Fig.7 in unit of cpm/m^2 as well as in unit of c/m^2. When the exposure period is longer than a few days, the efficiency of collecting the radioactive dust appears to drop considerably. In 1955, the radioactivity of the dust collected by the greased paper was measured 46 times and the total number of days of exposure was 225 days with the average exposure period of 4.89 days or roughly about 5 days. The total of the grossactivity detected from the exposed paper was about 5.11×10^{-8} curies/m^2 with the average grossactivity per one day of exposure about 2.27×10^{-10} curies/m^2-day. The total of the grossactivity detected from the rain collected during the period

from Feb. 16 to Dec.31, 1955 was about 1.58×10^{-7} curies/m^2. (Fig.7)

Therefore, the total of the grossactivity detected by exposing the greased paper to air may be estimated to be roughly about 32% of the total grossactivity detected from the rain with the average exposure period of about 5 days in 1955. In 1956, the radioactivity of the dust collected by the greased paper was measured 175 times and the total number of days of exposure was 352 with the average exposure period of about 2.01 days or roughly about 2 days. The total of the grossactivity detected from the exposed paper was about 3.46×10^{-7} curies/m^2 with the average grossactivity per one day of exposure about 9.82×10^{-10} curies/m^2.

The total of the grossactivity detected from the rain was about 4.91×10^{-7} curies/m^2 in 1956. Therefore, the total of the grossactivity detected by exposing the greased paper to the air may be estimated to be roughly about 71% of the total grossactivity detected from the rain with the average exposure period of about 2 days in 1956.

In 1957, the radioactivity of the dust collected by exposing the greased paper to the air was measured 155 times and the total number of days of exposure was 285 days with the average exposure period of 1.84 days or roughly about 2 days. The total of the grossactivity detected from the exposed paper was about 1.92×10^{-7} curies/m^2 with the average grossactivity per one day of exposure about 6.73×10^{-10} curies/m^2-day. The total of the grossactivity detected from the rain was about 3.67×10^{-7} curies/m^2 in 1957. Therefore, the total of the grossactivity detected by exposing the greased paper to the air may be estimated to be roughly about 54% of the total grossactivity detected from the rain. However, since the total number of days of exposure 285 days corresponds

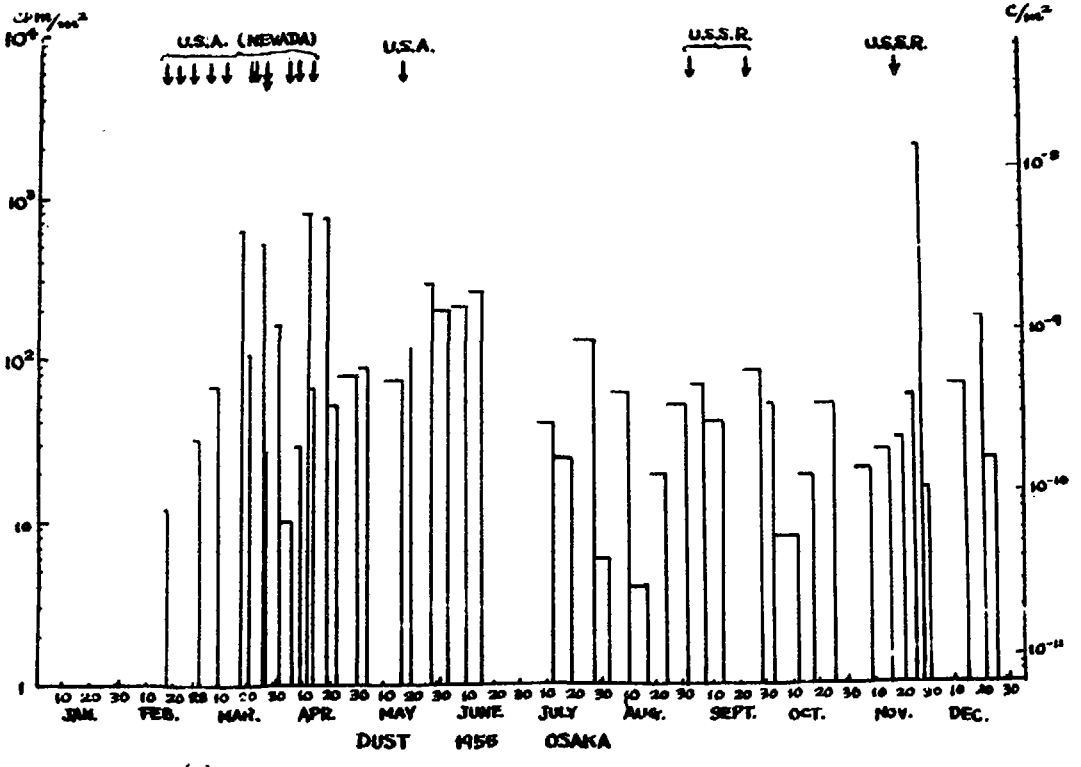


Fig. 7(a) The length of the horizontal bar indicates the number of days of exposure of the greased paper to the air.

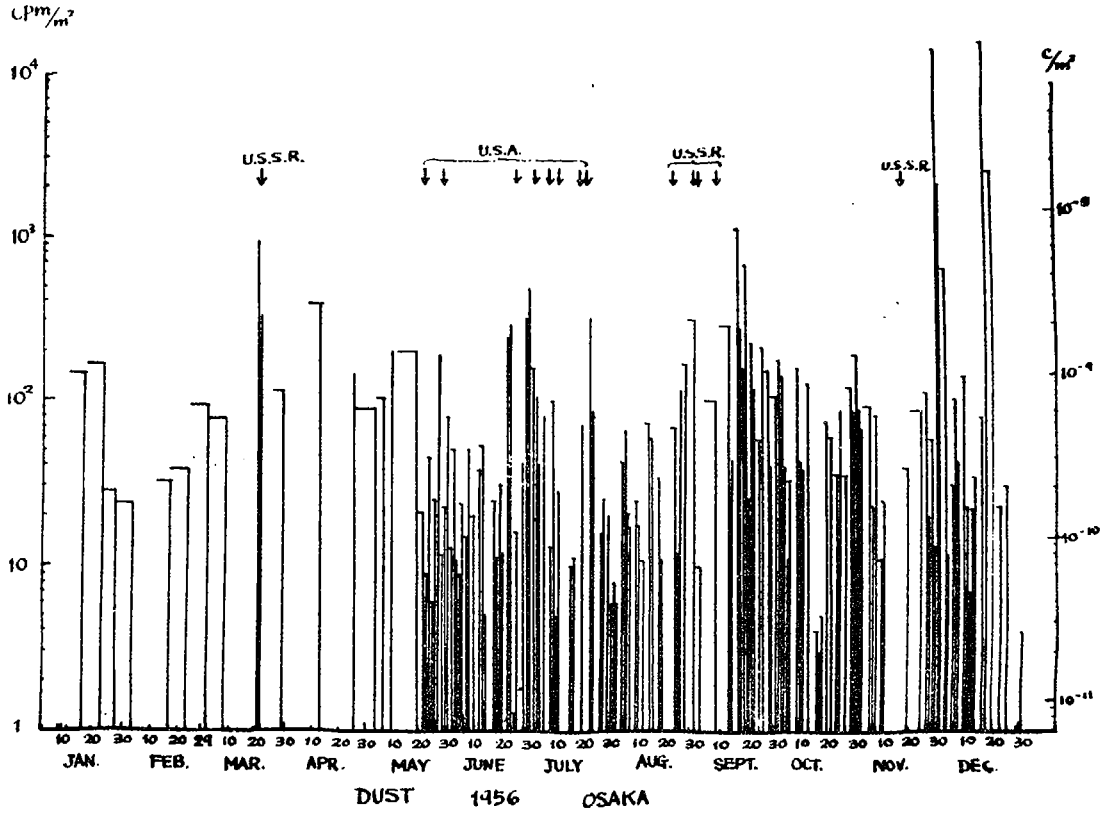


Fig. 7(b) The length of the horizontal bar indicates the number of days of exposure of the greased paper to the air.

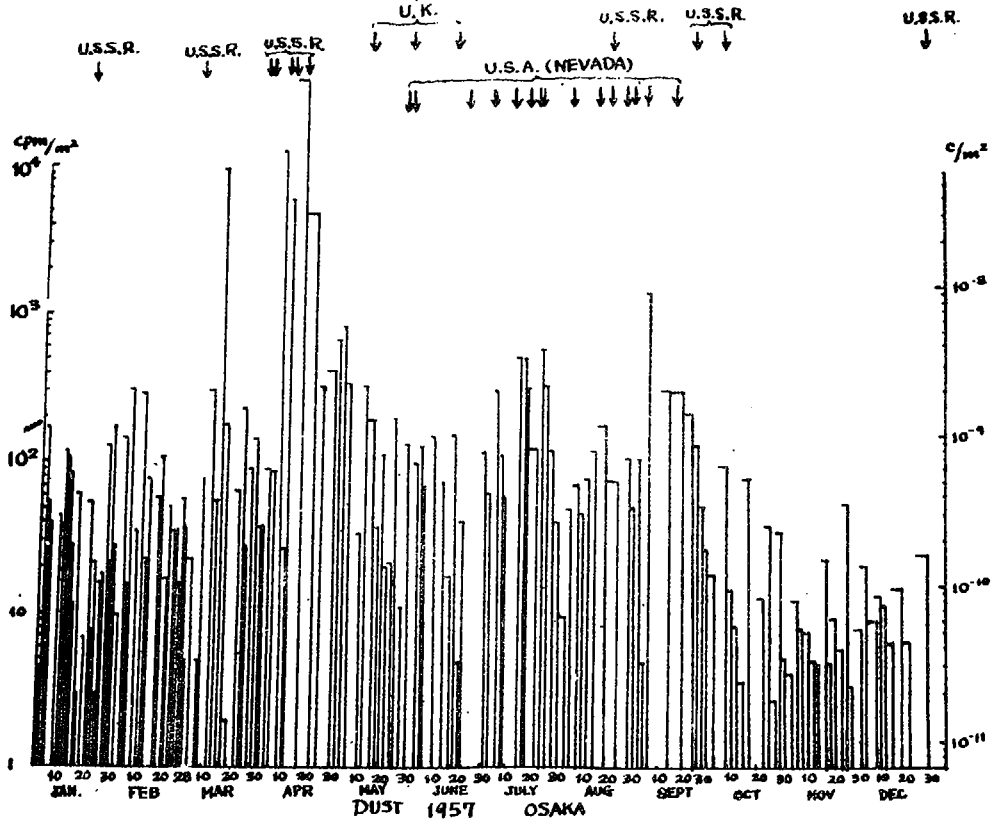


Fig. 7(c) The length of the horizontal bar indicates the number of days of exposure of the greased paper to the air.

to about 78% of a year, if we assume about 78% of the total grossactivity detected with the rain per year may correspond to the total grossactivity for 285 days, the percentage of the grossactivity detected by the exposed paper to that by the rain may be estimated to be roughly about 70% with the average exposure period of about 2 days in 1957. This percentage is roughly about the same order of magnitude as that of the previous year.

The frequency distribution of the radioactive deposition as detected from the exposed paper is shown in Tables 10 and 11. As can be seen in these tables, the radioactive deposition was clearly observed to have been increasing during the period from 1955 to 1957.

(Table 10)
(Table 11)

From the above analysis, it may be estimated that the over all efficiency of detection of radioactivity by exposing the greased paper to the air with the average exposure period of one to two days is roughly about 70% as compared with the grossactivity detected by the method of collecting the rainwater. However, the efficiency of detection of grossactivity appears to fall down to roughly about 30% with the average exposure period of about five days as can be seen from the comparison of the data obtained in 1955 with our method of detection.

The monthly totals of the grossactivity detected by the exposure of greased paper to the air during the period from Feb. 1955 to Dec. 1957 are summarized in Fig.8.

(Fig.8)

The infinite plane gamma ray exposure dose at a height of about one meter during a 30-year period from the total radioactive fallout as measured by the method of collecting the rainwater during

Table 10 : Dust

C/m² / cpm/m² / No. and % / Year	$< 6 \times 10^{-11}$		$6 \times 10^{-11} \sim 3 \times 10^{-10}$		$3 \times 10^{-10} \sim 6 \times 10^{-10}$		$6 \times 10^{-10} \sim 6 \times 10^{-9}$		$6 \times 10^{-9} <$		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 <			
	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
1955	4	8.7	14	30.4	14	30.4	13	28.3	1	2.2	46	100
1956	40	22.8	63	36.0	25	14.3	42	24.0	5	2.9	175	100
1957	31	20.0	47	30.3	25	16.1	46	29.7	6	3.9	155	100

Table 11 : Dust

Year \ Ci/m² day, cpm/m² day, No and %	< 6 × 10⁻¹¹		6 × 10⁻¹¹ ~ 3 × 10⁻¹⁰		3 × 10⁻¹⁰ ~ 6 × 10⁻¹⁰		6 × 10⁻¹⁰ ~ 6 × 10⁻⁹		6 × 10⁻⁹ <		Total	
	0 ~ 10		10 ~ 50		50 ~ 100		100 ~ 1000		1000 <			
	No.	%	No.	%	No.	%	No.	%	No.	%	No.	%
1955	19	41.4	17	37.0	5	10.8	5	10.8	0	—	46	100
1956	47	26.9	72	40.0	26	14.9	26	14.9	4	2.3	175	100
1957	19	12.3	64	40.3	34	21.9	34	21.9	4	2.6	155	100

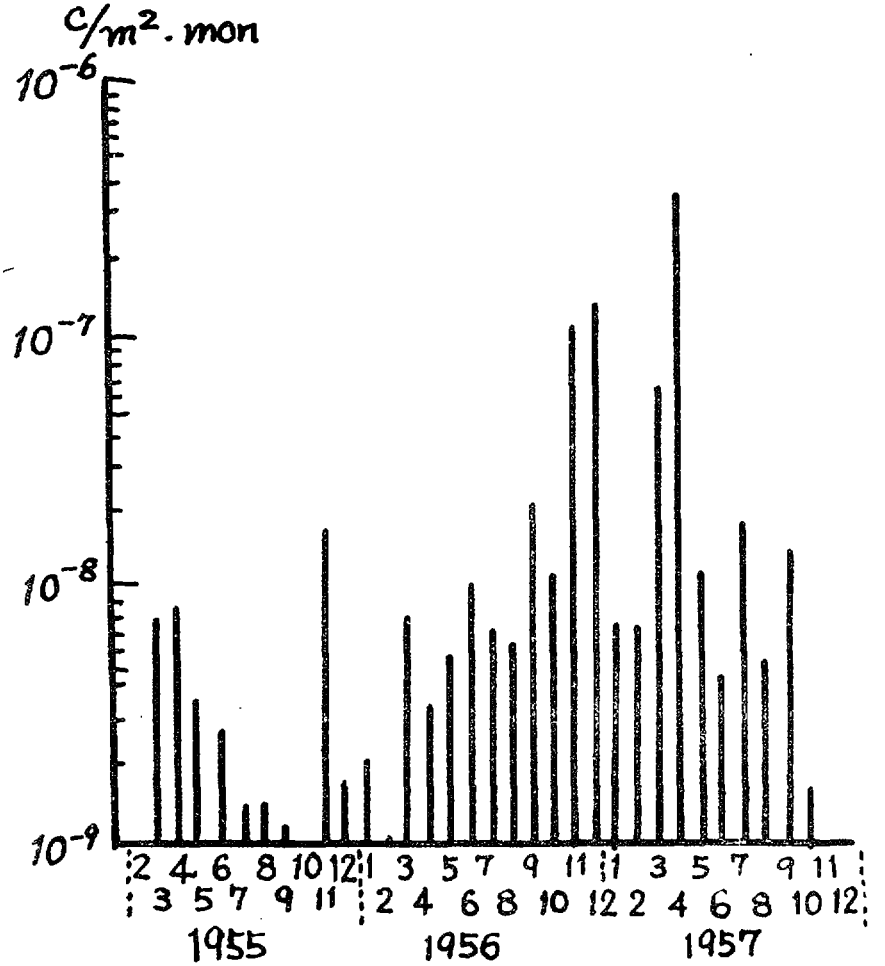


Fig. 8 : DUST OSAKA

the period from April 1954 to the end of August 1961 may be estimated to be roughly about 109 mrad with the method described in the previous paper[11] as a total sum of the estimates based upon the effective of the grossactivity of each rain.

However, if we assume double the total radioactivity estimated from our measurements would be the representative value for Japan as discussed in the above, the infinite plane gamma ray exposure dose during a 30-year period may be estimated to be about 218 mrad. Since there may be a difference by a factor of nearly two in the measurements of radioactivity depending on the location and the method of collection of rainwater, the 30 year dose in Osaka district may be estimated to be roughly in a range of about 109 - 218 mrad.

(III) Discussion and Summary

In the case of the American and British tests in the Pacific or the Soviet tests in the Arctic or Central Asia, the first circuit of radioactivity was usually observed to arrive in Japan a few days to about a week after the nuclear detonation. These radioactivies observed in the first circuit may be ascribed mostly to the tropospheric fallout with the apparent initial half decay period of the order of about a few days. The effects of the American tests in Nevada and of the French tests in Sahara were observed in Osaka about 10 -20 days after the nuclear tests, but the British tests in Australia in the Southern hemisphere could hardly be distinguished from the radioactivity measurements of the dust included in the rain in Japan.

This might probably be due to the fact that the diffusion of the radioactive dust across the stable air barrier between the Southern and the Northern Hemisphere is slow and is not likely to reach significant proportions in the time required for a major fraction of the dust in the troposhere to be deposited. Therefore the part of the radioactivity that spread in the troposphere at the time of nuclear detonation may be considered to be confined mostly to the hemisphere in which the nuclear tests were conducted, although the part of the fine radioactive dust that was injected high up into the stratosphere may be considered to be distributed all over the entire earth's surface.

The radioactivity of the rain was measured 668 times during the period from April 1954 to the end of August 1961. The annual total of the grossactivity detected per unit area by the method of collecting rainwater was 2.29×10^{-7} c/m^2 in 1954, 1.64×10^{-7} c/m^2 in 1955, 4.94×10^{-7} c/m^2 in 1956, 3.67×10^{-7} c/m^2 in 1957, 3.01×10^{-6} c/m^2 in 1958, 1.064×10^{-6} c/m^2 in 1959, 1.90×10^{-7} c/m^2 in 1960, and 2.37×10^{-8} c/m^2 in 1961, or roughly about 5.54 μc/m^2 in total during the period from April 1954 to the end of August 1961.

The annual average concentration of rainwater was estimated to be 2.81×10^{-4} μc/l in 1954, 1.55×10^{-4} μc/l in 1955, $3.34 \times$

10^{-4} μc/l in 1956, 2.54 x 10^{-4} μc/l in 1957, 3.2 x 10^{-3} μc/l in 1958, 6.56 x 10^{-4} μc/l in 1959, 1.73 x 10^{-4} μc/l in 1960, and 3.65 x 10^{-5} μc/l up to the end of August 1961.

There was a general tendency that the frequency of higher concentration was higher with the smaller amount of rainfall, while that of lower concentration higher with the larger amount of rainfall.

From the comparison of the data in 1960 and those in 1961 the half deposition time of the relatively longer lived components of the gross activity was estimated to be about 400 - 800 days or roughly about 2 years with the correction for the possible effects due to French tests.

Multiplying the total grossactivity per unit area 5.54 μc/m^2 by the total area of Japan, the total radioactivity carried over to Japan during the period of observation may be estimated to be roughly about two to three million curies.

The grossactivity of the dust collected on greased paper was also measured 376 times during the period from Feb. 1955 to the end of Dec. 1957. Comparing with the grossactivity detected by the method of collecting rainwater, the efficiency of detection of radioactivity by exposing the greased paper to the air was estimated to be about 30% with the average period of a single exposure about 5 days in 1955, and about 70% with the average period of exposure of about 2 days in 1956 and in 1957.

At the open place where there would be no hindrance in collecting the highly radioactive initial rainfall and at the closed place surrounded by the trees or buildings, it was not unusual to find a difference by a factor of more than two in the amount of collected radioactivity. This difference was especially large when it was windy. Depending upon the sampling location and method it was

oftentimes experienced to find a difference by a factor of two or three in the amount of detected radioactivity even in the same district. Therefore, it is not surprising to find a difference by a factor of nearly two or so in the amount of monthly deposition of Sr^{90} reported by the Japan Meteorological Agency[8] and that by Izawa,[11] et al for Tokyo district, although the two sampling places are only a few kilometers apart in the City of Tokyo.

The cumulative deposition of strontium-90 and cesium-137 at the end of August 1961 was estimated to be about 17.3 mc/km^2 of Sr^{90} and 33.64 - 43.6 mc/km^2 of Cs^{137} respectively, assuming the deposition of strontium-90 and cesium-137 in March 1954 to be about 0.5 mc/km^2 in Osaka district. The total amount of Sr^{90} carried over to Japan during about seven and a half years period of observation may be estimated to be roughly about 6×10^3 - 1.2×10^4 curies, while that of Cs^{137} about 1.2×10^4 - 3.1×10^4 curies.

Although the ratio of cesium-137 to strontium-90 was estimated to be about 1.7 - 2.2 prior to 1957, the over all ratio during the period of observation was estimated to be roughly about 2 - 2.6. From the analysis conducted during the period from spring to summer in 1958 the ratio of cesium-137 to strontium-90 was estimated to very from 1.7 to 3.3 with the average ratio of about 3.0 during this period.

According to the monthly measurements conducted by Izawa, et al during the period from August 1957 to June 1960 in Tokyo district (Tokyo and Chiba), the ratio Cs^{137}/Sr^{90} was observed to vary from 1.7 to 8.2 with the average ratio of about 2.8 during this period. Even in Tokyo and in Chiba which is an adjacent city to Tokyo, the ratio Cs^{137}/Sr^{90} was sometimes observed to be different by a factor of about two even in the same month and even with the same method of analysis conducted by the same person.

Judging from these observations, there appears to be a considerable seasonal as well as local variations in the ratio of cesium-137 to strontium-90 in the fallout. Therefore the overall ratio of cesium-137 to strontium-90 in Japan may be assumed to be roughly in the range of about two to three on an average.

The reason for such a wide variation in the ratio of cesium-137 to strontium-90 is not exactly known. The yield of cesium-137 and strontium-90 may be different depending on the type of nuclear weapons. The fraction of strontium-90 and cesium-137 escaping the local fallout and transported to a remote place may be different depending on the type and condition of nuclear detonation. The longer physical half life of inert gas parent of cesium-137 as compared with that of strontium-90 may cause the ratio Cs^{137}/Sr^{90} tend to be higher at the remote places than in the local fallout under certain conditions, although what may be considered chemically inert gas may be in a highly active state with most of the orbital electrons stripped off at the time of nuclear detonations.. There may be various meteorological factors, which may cause some fractionation or selection of certain nuclides during transportation of a mixture of radioactive substances from the site of detonation to the site of sampling.

However, it is rather difficult to explain the local variation in the adjacent cities like Tokyo and Chiba during the same period of observation with the same method of analysis. One of the possible causes for such a variation might be due to the difference in the chemical and physical nature of the local industrial dust and fumes as well as of the naturally occurring dust or chemical components included in the rain which may be different locally as well as seasonally. It may also depend on the method of sampling, what kind of vessel we may use for collecting rainwater, or whether the particulate dust included in the rainwater would be completely filtered off

prior to the chemical analysis or whether only an aliquot of supernatant water will be used for analysis without completely filtering or without complete stirring of the sedimented dust, or whether the analysis would be made with the rainwater including dust sampled from the bottom of the rainwater collecting vessel. Depending upon the chemical and physical nature of the vessel walls and the dust included in the rain there may be a possibility of fractional adsorption of certain nuclides and depending upon the chemical components of the rain this degree of fractionation may be affected considerably. Especially when the rainwater is sampled in the industrial area or near the chemical reserch institute, we must be careful about the nature of the dust and fumes of various types, not to speak of the dusts and vapours of the various radioactive substances occasionally used in the laboratory or factory. Whether the hydrochloric acid gas or nitric acid gas or other various types of inorganic gas or dusts are coming out of the draft chimney of the nearby laboratory or factory, or whether various types of organic gas or dusts are being exhausted from the chimney during the period of collection of rain water might cause the difference in degree of fractionation depending on the methods of sampling prior to the chemical analysis. Even in Osaka district, the decay rate of grossactivity of the rain collected on the same day at the Osaka City University and at the Osaka Prefectural Central Radiation Laboratory in the suburb of Sakai City which is adjacent to Osaka was occasionally observed to be considerably different after the French test in Feb. 1960 in spite of the fact that the method of measurement of radioactivity was very much the same.

In the country districts sometimes a considerable amount of pollen is included in the rainwater sample in spring besides the famamous yellow sand from the continental China.

In view of such difficulties in the standardization of the location and method of collecting and sampling the rainwater our data should also be regarded as the one that would give an order of magnitude estimation of the radioactive fallout for Osaka district and the difference in the total radioactive fallout by a factor of less than about two detected in the past at different sites may not be regarded as the essential difference for the different districts or areas where the sampling sites are located.

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Studies on the Radioactive Contamination due to Nuclear Detonations V

(The Radioactive Contamination of Human Body by Sr^{90} and Cs^{137} and its Corelation with the Fallout rate and Ground Deposition)

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The Radioactive Contamination of Human Body by Sr^{90} and Cs^{137} and its Corelation with the Fallout Rate and Ground Deposition

(I) Concentration of Sr^{90}

The radioactive contamination of human bone by Sr^{90} was measured with about 60 samples of human bone during the period from Jan. 1957 to July 1959. The results of these analysis have been reported to the United Nations Scientific Committee on the Effects of Atomic Radiation through Prof. Hiyama of Tokyo University.

(1) Method of Analysis

The sample of human bone was first ashed at about 900°C for about 4 hours in the electric furnace. The ashed bone was then pulverized and put in the centrifuge tube, about 5 gm of the sample in each tube with the capacity of about 250 ml. The amount of ashed bone used for the analysis was about 10 - 20 gm. Sr^{90} was separated as strontium nitrate after the addition of Sr-carrier from 75% nitric acid solution.(1)(2) After waiting for about 14 - 20 days for the growth of Y^{90}, Y^{90} was separated from Sr^{90} according to the ion exchange method described in the previous paper.(2) An example of the decay curve of the beta activity of yttrium fraction separated in this way is shown in Fig.1.

(Fig.1)

The activity of Sr^{90} was estimated from the beta activity of Y^{90} by extrapolating back to the time of separation from Sr^{90}. The existence of Y^{90} was confirmed by the half-line of approximately 64 hours. The beta activity was measured by the 2π-counter, 4π-counter as well as by the low back ground beta ray counter.

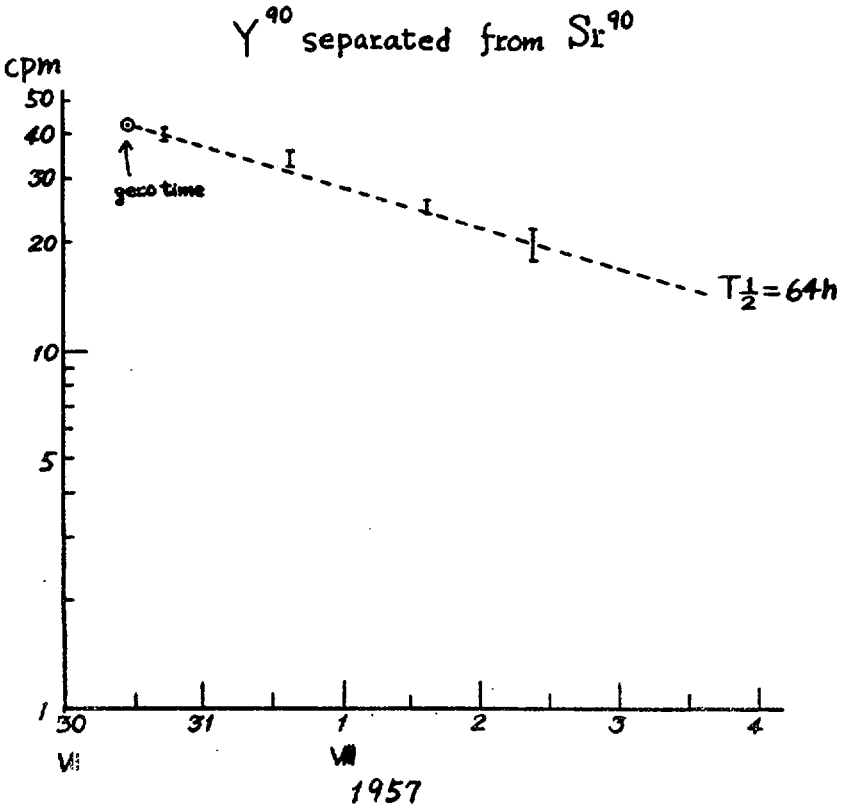
Y 90 separated from Sr 90
cpm
50
40
30
20
10
5
1
geco time
T½=64h
30
31
1
2
3
4
VI
VII
1957

Fig. 1

An aliquot of each sample was reserved for the determination of calcium.[3] The calcium is precipitated as calcium oxalate, which is insoluble in alkaline solution, by adding a soluble oxalate to the solution of the calcium salt. The precipitate is filtered, and dissolved in dilute sulphuric acid. Free oxalic acid is formed, which is titrated with a standard solution of potassium permanganate. The concentration of Sr^{90} is expressed in strontium unit (S.U.) which corresponds to 1μμc of Sr^{90} per 1 gram of calcium. The human bone of 70 kg standard man is assumed to contain about 1 kg of calcium.[8]

(2) Results and Discussion

The results of measurements of Sr^{90} are summarized in Table I - III, together with the possible statistical error estimated

(Table I)
(Table II)
(Table III)

from a series of measurements of activity. The average concentrations of Sr^{90} estimated for different age groups are shown in Table IV and Fig.2. As can be seen in the table, as a general tendency the average concentration of Sr^{90} in the bones of the children younger than 10 years is higher than that of the adults and the average concentration for the age group of 5 to 10 years is more than three times higher than that for the adults order than 30 years.

(Table IV)
(Fig.2)

This might probably be due to the fact that the children's bone is now being actively made of the calcium with higher concentration of Sr^{90}, while the adults' bone is already made of the old calcium with lower concentration of Sr^{90}.

Sr^{90} Concentration of Human Bones (From Jan.1957 to the end of 1957)

Table I

No.	Sex	Age at Death	Date	Bone	Ca/g ash	Sr^{90}/g ca	Cause of Death
1 O14	F	2 2/3	1957 1	Mixture of different bones	0.39	1.46 ± 0.11	myeloic Leukemia
2 # 2	F	19	" 2	Femur	0.36	0.34 ± 0.08	Nephritis
3 O 1	F	1 7/12	" 3	"	0.36	0.35 ± 0.12	Pneumonia
4 O 2	M	23 1/2	" 3	"	0.38	1.01 ± 0.11	lobar Pneumonia
5 O 5	M	24 1/3	" 5	"	0.40	0.73 ± 0.17	Soponific Poisoning
6 O 6	M	0	" 5	Mixture of different bones	0.37	0.04 ± 0.10	Stillbirth (Preg. X g.m.)
7 O 7	F	82	" 5	Femur	0.40	0.71 ± 0.12	Gastroenteritis
8 O 8	F	41	" 5	"	0.35	0.35 ± 0.13	Cancer of the breast
9 # 3	M	3 5/12	" 5	long Bones, Rib, Vertebral column	0.36	0.85 ± 0.23	Gastritis
10 O 9	F	79	" 6	Femur	0.40	0.25 ± 0.17	Nephrosklerosis
11 O10	F	5	" 6	"	0.40	1.21 ± 0.17	Asphyxia
12 O11	F	7/12	" 6	Femur, Vertebral column	0.37	0.81 ± 0.08	Measles
13 O12	F	30	" 6	"	0.38	0.25 ± 0.13	Heart Disease
14 O13	M	28	" 6	Femur	0.39	0.05 ± 0.05	Hemiplegia
15 O16	M	7 11/12	" 6	Rib, Vertebral column	0.38	1.75 ± 0.11	Asphyxia
16 O17	F	19 5/12	" 6	Rib	0.37	0.35 ± 0.10	Heart failure
17 O18	M	59	" 6	Skull, Femur	0.38	0.23 ± 0.07	Sepsis
18 O19	F	8 5/12	" 6	Rib, Skull	0.38	0.20 ± 0.08	Meningitis
19 O20	M	46	" 6	Rib, Vertebral column	0.36	0.07 ± 0.05	Heast failure
20 # 5	M	11/12	" 7	Mixture of different bones	0.38	0.65 ± 0.13	Encepharitis
21 # 6	M	70	" 7	Femur	0.37	0.14 ± 0.05	Cancer of the Stomach
22 OF1	M F F	preg.VII, V, V, g.m.	" 4,5, 6	Mixture of different bones	0.37	0.02 ± 0.08	Abortion
23 OF2	M M M	preg.VI, VIII, VI, g.m.	" 7,7, 8	"	0.37	0.35 ± 0.09	"
24 OF3	M F F M	prog.VII, VI, VI, VIII, g.m.	" 8,9, 10,10	"	0.38	0.41 ± 0.09	"

O:Osaka, W:Wakayama, OF:Fetus in Osaka, Preg.= Pregnancy, g.m. = gravidal month

Sr^{90} Concentration of Human Bones (From Jan.1958 to Dec.1958)

Table II

No.	Sex	Age at Death	Date	Bone	Ca/g ash	Sr^{90}/g ca	Cause of Death
25 022	F	1/12	1958 1	Mixture of different bones	0.37	1.70 ± 0.17	Pneumonia
26 023	F	1/4	" 1	"	0.37	0.28 ± 0.09	Meningitis
27 024	M	0	" 1	"	0.38	0.02 ± 0.09	Stillbirth (Preg. X g.m.)
28 025	M	57	" 1	long Bones, Rib, Vertebral column	0.36	0.23 ± 0.10	Chronic Alcoholism
29 026	F	57	" 1	long Bones, Vertebral column	0.38	0.12 ± 0.08	Cancer of the liver
30							
30 031	M	13 1/6	" 6	Femur	0.36	0.51 ± 0.09	Panmyelophthisis
31 032	F	4 1/3	" 6	"	0.37	0.38 ± 0.09	Acute myeloic Leukemia
32 033	F	4 11/12	" 6	"	0.36	0.86 ± 0.09	Acute Enteritis
33 034 F F F		0	" 6	Mixture of different bones	0.36	0.13 ± 0.09	Premature
34 035	F	1/4	" 6	"	0.37	0.83 ± 0.10	Pylorus Stenosis
35 037	M	16 7/12	" 6	Femur	0.37	0.27 ± 0.07	Acute Pneumonia
36 038	M	4 1/6	" 6	"	0.37	0.57 ± 0.09	Heart failure
37 039	F	2 2/3	" 6	Mixture of different bones	0.37	0.90 ± 0.09	respiratoric paralysis
38 040	M	11 1/3	" 6	"	0.37	0.28 ± 0.08	Peritonitis carcinomatosa
39 041	M M	0	" 6	"	0.38	0.07 ± 0.08	Stillbirth (Preg. X g.m.)
40 042	F	1/4	" 6	long Bones, Rib, Vertebral column	0.37	0.35 ± 0.08	Pneumonia
41 043	M	0	" 6	Mixture of different bones	0.36	0.42 ± 0.19	Premature
42 045	M	14 11/12	" 6	Femur	0.37	0.08 ± 0.11	Asphyxia
43 047	M	18 1/4	" 6	Rib, Tibia	0.37	0.47 ± 0.09	Heart failure
44 048	M M	0	" 6	Mixture of different bones	0.37	0.56 ± 0.09	Stillbirth (Preg. X g.m.)
45 050	F	5 0/12	" 6	Femur	0.37	0.75 ± 0.09	Enteritis
46 051	F	7/12	" 6	Mixture of different bones	0.38	1.10 ± 0.08	Pneumonia
47 060	M	11 5/12	" 6	Femur	0.37	0.30 ± 0.07	Cardiac Asthma
48 061	F	15 0/12	" 6	"	0.36	1.47 ± 0.08	Heart failure
49 044	F	2 5/12	" 7	Mixture of different bones	0.36	0.25 ± 0.11	" "
50 046	M	13 1/3	" 7	Femur	0.36	0.75 ± 0.09	Leukemia
51 059	M	6 2/3	" 7	"	0.37	1.29 ± 0.08	Pneumonia
52 052 M M F		0	" 12	Mixture of different bones	0.37	0.12 ± 0.07	Premature
53 053	F	2 2/3	" 12	long Bones, Rib	0.36	1.04 ± 0.07	Fracture of the skull
54 054	M	3/4	" 12	Mixture of different bones	0.37	0.17 ± 0.07	Dyspepsia

Sr^{90} Concentration of Human Bones (From Jan.1959 to Jnl.1959)

Table III

No.	Sex	Age at Death	Date		Bone	Ca/g ash	Sr^{90}/g ca	Cause of Death
55 062	M	1 3/12	1959	1	Mixture of different bones	0.37	0.91 ± 0.08	Fracture of the skull
56 063	M	7/12	"	7	"	0.37	0.39 ± 0.08	measles Pneumonia
57 064	F	22 4/12	"	7	"	0.37	1.17 ± 0.09	Death by drawning
58 065	M	9 1/12	"	7	"	0.37	0.53 ± 0.07	Ekiri
59 066	M	1/12	"	7	"	0.37	0.59 ± 0.07	Ikterus neonatorum gravis

Table IV

Age	Year	Sr^{90} in Human Bone (S.U.)								Average	Total Average
Fetue Stillborn - 1M.	1957	0.02	0.04	0.35	0.41					0.205	
	1958	0.02	0.07	0.12	0.13	0.42	0.56			0.221	0.214 (10)
	1959										
1M. - 1Y.	1957	0.65	0.81							0.730	
	1958	0.17	0.28	0.35	0.82	0.83	1.10	1.70		0.751	0.699 (11)
	1959	0.39	0.59							0.490	
1Y. - 5Ys.	1957	0.35	0.85	1.21	1.45					0.968	
	1958	0.25	0.38	0.57	0.70	0.75	0.86	1.04		0.665	0.778 (12)
	1959	0.91								0.910	
5Ys. - 10Ys.	1957	0.20	1.75							0.975	
	1958	0.93	1.29							1.110	0.940 (5)
	1959	0.53								0.53	
10Ys. - 20Ys.	1957	0.34	0.35							0.345	
	1958	0.08	0.27	0.28	0.30	0.47	0.51	0.75	1.47	0.516	0.482 (10)
	1959										
20Ys. - 30Ys.	1957	0.05	0.25	0.73	1.01					0.510	
	1958										0.642 (5)
	1959	1.17								1.17	
30Ys. -	1957	0.07	0.14	0.23	0.25	0.35	0.71			0.291	
	1958	0.12	0.23							0.175	0.262 (8)
	1959										

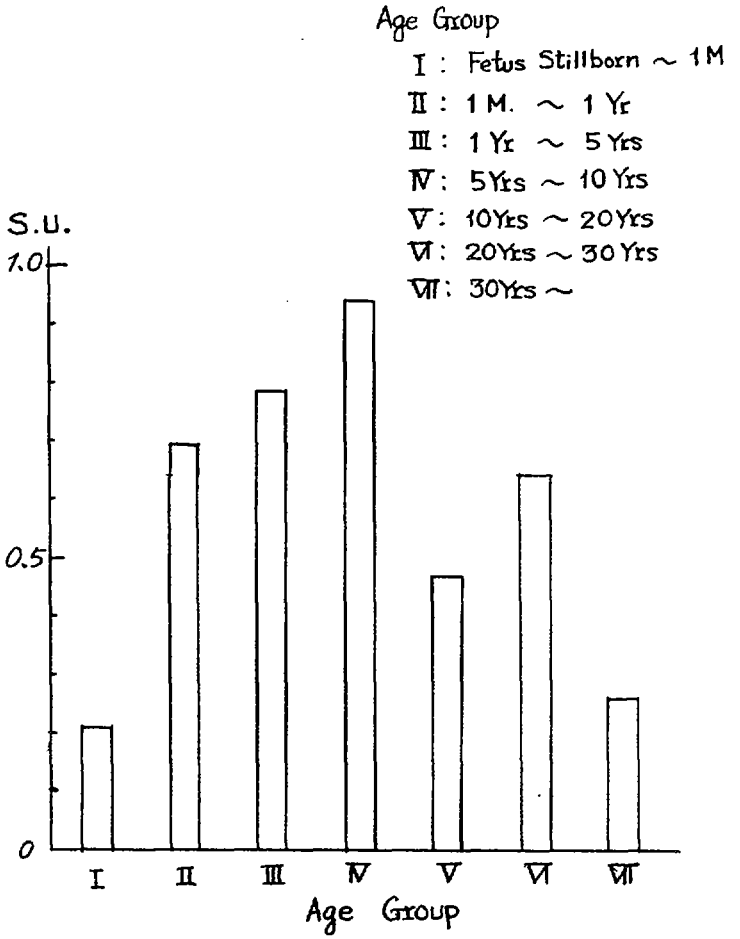

Fig.2: Concentration of Sr^{90} in Human Bone in Strontium Unit (Sr^{90} $\mu\mu c/g\text{-}Ca$)

The highest concentration during the period of observation is found among the children to be about 1.75 S.U.. Although the yield of chemical analysis was estimated to be about 60 - 80%, taking into consideration the possible range of error in the estimation of the yield in radiochemical analysis besides the statistical error estimated from a series of measurements of activity indicated in the table, this might be interpreted as to correspond to roughly about 2 S.U..

During the period of measurements, we have occasionally encountered a few cases with considerably higher concentration of Sr^{90}. The concentrations as high as 2.3 - 4.8 S.U. were detected in the fetus and the child, the bone samples of which were obtained from the crematory. However since the past history of such cases was not clear, the cases with unusually high concentration have not been reported.

In connection with this, it should also be noted that most of the cases examined in our laboratory and listed in the tables are pathological ones and the results may not be considered to be representative values for the average normal people and the average values shown in Fig.2, may be considered somewhat lower than the representative values for the average normal person.

The concentration of Sr^{90} in human bone may greatly depend on the food habit of the individuals. According to the white paper on the citizen's nutrition[4] published by the Public Health Division of Osaka City Government, the calcium source in the average diet may be assumed to be as follows: about 260 mg from cereals and vegetables, about 20 mg from milk and dairy products about 100 mg from fish and marine products or abput 380 mg in total on the average. In 1956, the radioactive

contamination by Sr^{90} was about 49 S.U. in polished white rice about 9.4 S.U. in vegetables, about 2.9 S.U. in milk and dairy products, about 0.29 S.U. in marine fish.(5) In 1957 and 1958, the radioactive contaminations of white rice and vegetables were about 24.2 and 56.4 S.U.. From these figures the average daily intake of Sr^{90} during the period of observation may be estimated at about 5 - 13.4 μμc.

In certain part of Osaka, it has been encouraged for the adults and children to take brown rice to maintain health because it is rich in vitamins and minerals such as calcium. However, the radioactive contamination of brown rice by Sr^{90} was reported to be unusually high during the period of observation, reaching as high as 81 - 250 S.U. The Sr^{90} concentration in the food served at the hospital may be much lower than that for the average healthy person, because only the well polished white rice is usually served for the patients besides various medicines with possibly much less contamination of Sr^{90}. Therefore, depending on the food habit and the dietary composition and whether people depend or not on the unfiltered rainwater as the only source of drinking water in the isolated small island or in the mountain district, or whether the person has been hospitalized for a long time or not, the concentration of Sr^{90} in human bone may be expected to be considerably different. Although the rate of transfer of Sr^{90} from the soil deposition to the human bone may also depend on the discrimination factor which is determined by the ratio of stable strontium to calcium in each member of the food chain, judging from what is discussed in the above it may not be too much to assume that the concentration of Sr^{90} in the bone of average normal children under the dietary composition and food habit of great variety as seen at present in Japan might reach 2 - 5 S.U. corresponding to the cumulative deposition of Sr^{90} of about 10 mc/km^2 and the fallout rate of about 3 mc/km^2 - year.(2)

(3) Effect of calcium content in the diet on the deposition of radioactive strontium in the bone.

In case of calcium deficiency, a greater percentage of administered Sr^{90} may be expected to deposit in the bone. In order to examine about this point, to three different groups of young rats weighing about 55 gm which were grown with the food of low calcium (0.15%), standard calcium (0.44%) and high calcium (1.03%) content respectively, 0.28 μc per one gram of body weight of carrier free strontium-89 in the form of strontium chloride was directly administered orally into the stomach of the rats with the special syringe. The radioactivity in different organs was examined after autopsy at different time intervals. The deposition of strontium-89 in the femur was observed to reach a maximum at 4 - 5 hours after administration of the radioactive strontium as shown in Fig.3. In the fighre each point represents the mean value of

(Fig.3)

three rats and the percentage is expressed with the percentage of the activity detected unit mass of the organ against the administered dose per one gram of body weight. As can be seen in the figure, at two days about 7% of the administered strontium was observed to deposit in the bone with the group of low calcium content, about 4.5% with the group of standard calcium content and about 2.5% with the group of high calcium content.(9)

Judging from these results, the deposition of radioactive strontium in the bone with the group fed with high calcium diet may be roughly about one half to one third of that of the low calcium group. Therefore if the calcium is supplemented with the radioactive free calcium pills, the rate of deposition of Sr^{90} in human bone may also be reduced considerably as pointed out by Prof. Linus Pauling of California Institute of Technology.(10)

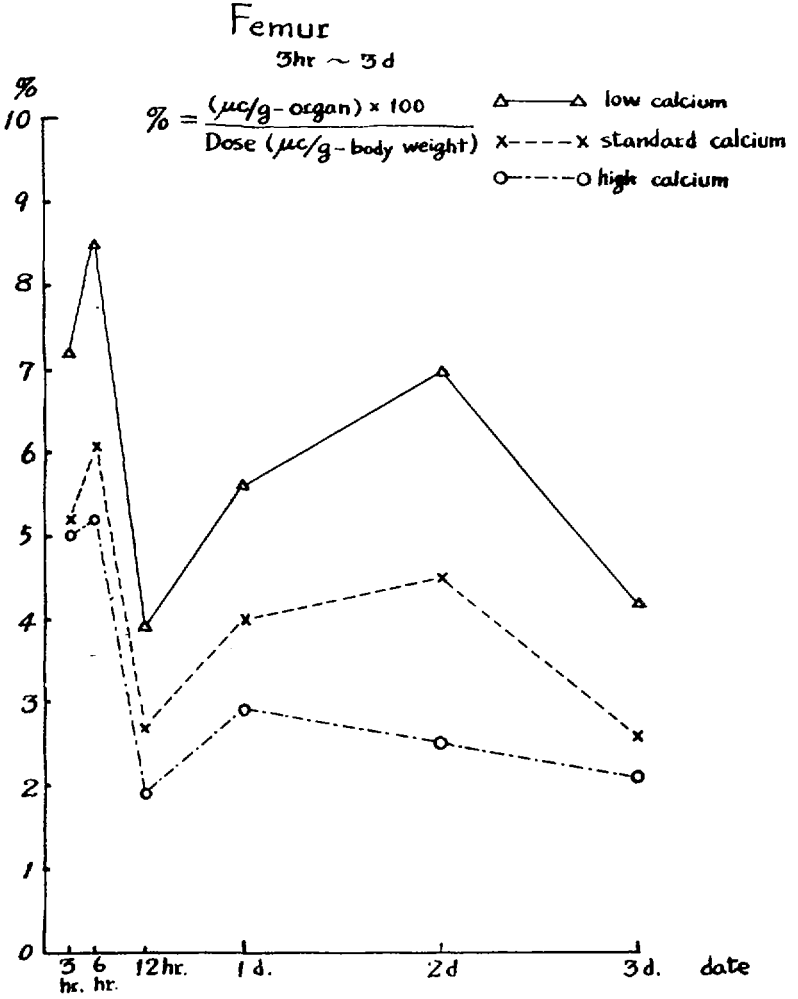
Femur
3hr ~ 3d
%
10
9
8
7
6
5
4
3
2
1
0
% = (μc/g-organ) × 100 / Dose (μc/g-body weight)
low calcium
standard calcium
high calcium
3 hr.
6 hr.
12hr.
1 d.
2d
3d.
date

Fig. 3

(II) Concentration of Cs^{137}

The radioactive contamination of meat and human muscle by the cesium-137 that has similar physical half-life as the strontium-90 was also measured in 1957. The cesium was separated as cesium bismuth iodide precipitate[11] from the alkaline metal fraction with carrier Cs after complete ashing of the sample. The radioactive free cesium carrier was kindly provided to the author from Prof. Nishi of Kyoto University. About 180 μμc of Cs^{137} per one kg of beef without fat was detected from the beef bought at the City market in Osaka in June 1957 after careful elimination of the fat part of the meat. In 1957 we have also examined uterus and ovary excised from the patients of the uterus myoma and the ovarian cystoma at the Department of Gynecology of the Osaka City University Hospital and detected about 100 μμc per 1 kg of muscle. From the analysis of the standard diet the average daily intake of Cs^{137} in urban district was estimated to be roughly about 50 μμc in 1957.[12]

The concentration of Cs^{137} in various food such as meat and vegetables in 1957 was estimated to be in the range of about 20 ~ 100 cesium unit, while that in human body about 30 ~ 100 cesium unit. One cesium unit corresponds to 1 μμc per 1 gram of potassium.[8][12] Assuming the amount of potassium in human body to be about 140 gram per 70 kg standard man, the total activity of Cs^{137} in human body may be estimated to be about 4.2 ~ 14 mμc corresponding to the cumulative deposition of about 14 mc/km^2 and the fallout rate of about 4 mc/km^2 ~ year.[2] However, in 1958 the concentration of Cs^{137} in human body in cesium unit has been reported to be about two to three times higher on the average in Tokyo than the above value.[5][6][15]

Some of the unusually high cases with the concentration of Cs^{137} in cesium unit in some of the organs of the human body higher by about 10 ~ 100 times or more than the above value in 1957 have been reported by Yamagata[5][15] in Tokyo for the sample obtained in the latter half of 1958. (A case with 1.86×10^4 cesium unit in prostate in Tokyo in Aug. 1958)[15]

(III) Discussion and Summary

About 60 bone samples were analysed for Sr^{90} during the period from Jan. 1957 to July 1959. The highest average concentration was found to be about 1 S.U. for the age group of 5 - 10 years old. However, since most of these cases are more or less pathological ones after long period of hospitalization the results may not be considered to be the representative values for the average normal person. The highest concentration among the cases whose past history is known was 1.75 S.U. or roughly about 2 S.U..

During the period of observation occasionally we have found higher concentration than 2 S.U. in the bone samples of a few cases obtained from the crematory. However, since the past history of these cases is not known, it is not clear whether they depended on the rainwater or on the brown rice with much higher contamination than the white rice.

In view of these findings it may not be too much to assume that the concentration of Sr^{90} in the bones of the average normal children might reach 2 - 5 S.U, corresponding to the cumulative ground deposition of about 10 mc/km^2 and the fallout rate of about 3 mc/km^2 - year.(2)

The concentration of Cs^{137} whose physical half-life is very much similar to Sr^{90} was in the range of about 30 - 100 cesium unit in 1957 to early 1958 corresponding to the cumulative deposition of about 14 mc/km^2 and the fallout rate of about 4 mc/km^2 - year.(2)

However, during the period from 1958 to early 1959 the concentration of Cs^{137} in some of the organs of the human body has been reported to be about two to three times higher on the average in Tokyo than the above value in 1957.

The concentration of Sr^{90} and Cs^{137} in human body may not only depend on the food such as meat and vegetables but also on the degree

of contamination of the drinking water and the air inhaled. In case of those who depend on the rainwater without adequate filtering of Sr^{90} and Cs^{137}, the major route of contamination of the human body may be assumed to be the rainwater used for drinking and daily cooking.

Although it is assumed in the UN report[8] that the concentration of Sr^{90} in human body depends only on the cumulative deposition while that of Cs^{137} only on the fallout rate, it seems to be more plausible to assume that the concentration of both Sr^{90} and Cs^{137} in human body might depend on the cumulative deposition and the fallout rate. The food habit of the people may be different not only locally and seasonally, but also with time. The food habit in the future may not remain the same as at present. The people who depend on the rainwater are now advised not to drink the rainwater without adequate filtering. Therefore it may be extremely difficult to predict the future level of Sr^{90} and Cs^{137} in human body, although the cumulative deposition and fallout rate might be predicted for the future.

The foliage retention of Sr^{90} by some of the plants and vegetables may not be negligible,[13] although the fraction of foliage absorption of Sr^{90} may not be as large as that of Cs^{137}. The root absorption of Cs^{137} by some of the plants and vegetables may not always be considered negligible, although the fraction of root absorption of Cs^{137} may not be as large as that of Sr^{90} and might depend greatly on the type of the plants, the nature of the soil and fertilizers and the degree of rainfall, irrigation, farming and ploughing, etc..

Taking these possibilities into consideration, it may be assumed that a total of roughly about 0.1 - 1 mμc of Sr^{90} in human body may correspond to the fallout rate of about 1 mc/km^2 - year and roughly about 0.1 - 0.2 mμc to the cumulative ground deposition of about 1 mc/km^2.

It may not be too unreasonable to assume that a difference by a factor of about ten in the degree of dependence of Sr^{90} level in human body on the fallout rate might exist between those who depend on well-polished white rice and the well-filtered tap water and those who depend on brown rice with high concentration of Sr^{90} and the rainwater without adequate filtering.

In case of the level of Cs^{137} in human body, the local, seosonal, and individual variation may be much larger than in case of Sr^{90} because the biological half life of Cs^{137} is much shorter than that of Sr^{90} in the bone. Therefore it may be rather difficult to estimate more accurately, because a difference of Cs^{137} level in various organs of the body by a factor as large as 10 - 100 may occasionally be found in various reports in Japan.(5)(15)

However, since the degree of foliage absorption of Cs^{137} by vegetables is assumed to be higher than that of Sr^{90} while the degree of root absorption of Cs^{137} lower than that of Sr^{90}, it may be assumed that the dependence of the level of Cs^{137} in human body on the fallout rate might be about the same as or about double the upper limit of Sr^{90} and the dependence of the level of Cs^{137} in human body on the cumulative ground deposition roughly about one half to one tenth that of Sr^{90}.

From these assumptions, it may be assumed for Cs^{137} that a total of about 1 - 2 mμc of Cs^{137} in human body may correspond to the fallout rate of roughly about 1 mc/km^2 - year and about 0.1 - 0.01 mμc to the cumulative ground deposition of roughly about 1 mc/km^2.

Because many uncertainties appear to be involved in the degree of root absorption of Cs^{137} by the plants or vegetables which might greatly depend on the nature of the soil, fertilizers and many other natural and artificial factors, it may not be too much to assume a

difference by a factor of about ten for the dependence of Cs^{137} level in human body on the cumulative ground deposition, although the major route of contamination of plants of vegetables by Cs^{137} might be through the foliage retention of Cs^{137} which depends on the fallout rate.

However, these corelations between the levels of Sr^{90} and Cs^{137} in human body and the cumulative ground deposition and the fallout rate should be revised in the future when more accurate quantitove data about the route of intake of such nuclides under natural conditions may become available. In order to estimate the average body burden of Sr^{90} and Cs^{137} in the population more accurately, more detailed informations not only about the discrimination factor expressed by the ratio of stable strontium to calcium or the ratio of stable cesium to potassium, but also about the fraction of people who depend on different food and different drinking water with different concentrations of Sr^{90} and Cs^{137} may be necessary.

Assuming the effective energy of Sr^{90} in the bone to be about $\Sigma EF(RBE)n=5.5$ MeV with the non-uniformity factor n=5,[7] the annual dose rate to the bone corresponding to 1 strontium unit or 1 mμc in 1 kg of calcium in the total bone of 7 kg may be estimated to be about 14.6 mrem per year, while with the non-uniformity factor n=1 it may be estimated at about 2.9 mrad per year. In case of Cs^{137} the non-uniformity factor is assumed to be n=1. Therefore, assuming the effective energy of Cs^{137} for the whole body to be about $\Sigma EF(RBE)n=0.59$ MeV, the annual dose rate corresponding to a continuous body burden of 1 mμc in 70 kg of whole body or the average concentration of roughly about 7.1 cesium unit in whole body may be estimated to be about 0.16 mrem or mrad per year.

In view of the various uncertainties involved in the radioecological behaviour of various nuclides, it may be desirable to keep the specific activity of each element in the human environment as well as in the human bocy below the level at which the total mean effective energy of the radiations emitted per unit time from the radioactive isotopes in one gram of the corresponding stable isotope would not exceed the rate of emission of the radiation energy equivalent to 0.01 rem/week.

At equilibrium state the energy generated per unit mass may be considered roughly about equivalent to the energy absorbed in it. Since 0.01 rem corresponds to 6.25×10^5 Mev/gram and one week to 6.05×10^5 seconds, the permissible specific activity (PSA) of the isotope may be expressed by the following equation.

$$[PSA] = \frac{(6.25 \times 10^5)}{(6.05 \times 10^5)\ \bar{E}_{eff}\ (3.7 \times 10^{-2})} \quad \mu\mu c/g\text{-element} \quad (1)$$

where $\bar{E}_{eff}$ is the mean effective energy per disintegration of the isotope in unit of Mev and may be assumed to be Σ EF(RBE). Simplifying the above equation we may obtain

$$[PSA] = \frac{28}{\bar{E}_{eff}} \quad \mu\mu c/g\text{-element} \quad (2)$$

At the disposal of the various radioactive nuclides it may be desirable to dilute the radioactive nuclides by the corresponding stable isotopes, down to the above level of permissible specific activity. In case of the radioactive nuclides without any corresponding suitable stable isotopes, it may be necessary to dilute the radioactive nuclide with the stable isotopes that have radioecologically and metabolically similar behavior to the

radioactive nuclide.

In case of Sr^{90}, $\Sigma EF(RBE)=1.13$ Mev estimated for the whole body may be taken as $\bar{E}_{eff}$. Substituting this value into the above equation we may obtain,

$$[PSA] = 25 \ \mu\mu c - Sr^{90}/g - Sr \qquad (3)$$

In case of Sr^{89}, since the mean energy of beta ray per disintegration may be assumed to be $\bar{E}_{eff}=0.56$ Mev,

$$[PSA] = 50 \ \mu\mu c - Sr^{89}/g - Sr \qquad (4)$$

In case of Ca^{45} with $\bar{E}_{eff}=0.086$ Mev,

$$[PSA] = 326 \ \mu\mu c - Ca^{45}/g - Ca \qquad (5)$$

In case of C^{14} with $\bar{E}_{eff}=0.054$ Mev,

$$[PSA] = 520 \ \mu\mu c - C^{14}/g - c \qquad (6)$$

Some of the results of these estimations of the permissible specific activity (PSA) are shown in Table V.

(Table V)

This was the idea proposed by Nishiwaki in 1954 at the time of Bikini Accident.[14] However, later the radioactive contamination of Sr^{90} in our environment was observed to increase seriously due to the repeated large scale nuclear testings.

If the specific activity of one isotope in our environment or in our body exceeds the permissible specific activity as defined in the above, the permissible specific activity of other isotopes that have radioecologically or metabolically similar behavior should be reduced correspondigly.

Table V ; Permissible Specific Activity

	$\overline{E}$ eff (MeV)	PSA ($\mu\mu c/g$)
H^3	0.01	2,800
C^{14}	0.054	520
P^{32}	0.69	41
Ca^{45}	0.086	326
Sr^{89}	0.56	50
Sr^{90}	1.13	25
I^{131}	0.44	65
Cs^{137}	0.59	48

The concentration of Sr^{90} during the period from 1956 to 1957 in Japan has been reported to be about 390 - 540 S.U. in rice bran and chaff, 153 - 170 S.U. in brown wheat and about 53 S.U. in wheat flour.(8) It seems to be apparent that the specific activity of Sr^{90} in stable strontium corresponding to the above concentration in S.U. may be higher by a factor of about 10^4 - 10^5 than the permissible specific activity of strontium discussed in the above.

Even if we assume a concentration of Sr^{90} in the bone to be about 1 S.U., assuming the amount of stable strontium in one gram of bone calcium to be about 10^{-4} gram, the specific activity of Sr^{90} in stable strontium may be estimated to be about 10^4 μμc-Sr^{90}/g-Sr which is about 400 times higher than the permissible specific activity of 25 μμc of Sr^{90}/g-Sr.

In such cases, the second permissible specific activity in our body may be considered to be 25 μμc of Sr^{90} per one gram of calcium which may correspond to 25 S.U.. However, if the concentration of Sr^{90} in calcium reaches this level, there would be no allowances left for calcium. Therefore, if the concentration of Sr^{90} in calcium approaches the second permissible specific activity the permissible specific activity of calcium should be reduced correspondingly. For instance, if the concentration of Sr^{90} in calcium becomes 12.5 μμc of Sr^{90}/g-calcium, the permissible specific activity of radioactive calcium should be reduced to one half. In case of Ca^{45} with PSA=326 μμc/g-Ca, the permissible specific activity should be reduced down to 163 μμc of Ca^{45}/g-Ca corresponding to the increase of Sr^{90} to a level of 12.5 μμc of Sr^{90}/g-Ca.

If the concentration of Sr^{90} in calcium exceeds the second permissible level of 25 μμc of Sr^{90}/g-Ca in the bone, the third permissible specific activity may be taken to be 25 μμc of Sr^{90}/g-bone. However, in such cases, the permissible specific activity of other radioactive nuclides such as H^3, C^{14}, P^{32} that have metabolically

similar behavior to other elements than calcium constituting the bone should also be reduced correspondingly.

The second permessible specific activity of 25 μμc of Sr^{90}/g-Ca may correspond to about 0.25 μc of Sr^{90}/g-Sr, assuming the concentration of stable strontium in the calcium of bone to be about 10^{-4} gram Sr/g-Ca, which is about 10^{4} times higher than the permissible specific activity of Sr^{90} in the stable strontium. The third permissible specific activity of 25 μμc of Sr^{90}/g-bone may correspond to 1.75 μc of Sr^{90}/g-Sr, assuming the fraction of calcium in the bone to be about 1/7, which is about 7×10^{4} times higher than the first permissible specific activity of 25 μμc of Sr^{90} per one gram of stable strontium.

The third permissible specific activity of 25 μμc of Sr^{90}/g-bone may correspond to the total burden of about 0.175 μc in the bone, which is roughly about 9% of the occupational maximum permissible total burden of 2 μc in the bone given in ICRP(1958). A total of 0.175 μc in the bone may be estimated to correspond to the annual dose rate of about 2.6 rems/year to the bone, assuming the non-uniformity factor of five for the bone. This level may be considered too high as the permissible level for the general public. The second permissible specific activity of 25 μμc of Sr^{90}/g-Ca or 25 S.U. may be estimated to correspond to the annual dose rate of about 0.37 rem/year to the bone.

It is desirable to keep the level of radioactive contamination in our environment below the level of the first permissible specific activity. However, because of the repeated nuclear testings in the past, the level of radioactive contamination of Sr^{90} in our environment has already exceeded even the level of the second permissible specific activity in some of the food such as the brown rice. Because of the discrimination factor of Sr^{90} which may be determined by the ratio of the stable strontium to calcium in the food chain, the concentration of Sr^{90} in calcium of human bone may be assumed to be

roughly about one tenth the level of the second permissible specific activity or less at present. However, with the increase of the cumulative deposition of Sr^{90} the level of Sr^{90} in human bone may have a tendency to increase gradually to reach the equilibrium value with the environmental contamination level.

Under such circumstances, we feel it extremely important to try to exert our utmost efforts not to have the concentration of Sr^{90} in human bone reach the level of second permissible specific activity in order to keep as much allowances as possible for other radioactive nuclides than the bomb-produced for the peaceful uses of atomic energy in our country.

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Studies on the Radioactive Contamination due to Nuclear Detonations VI

(Theoretical Analysis of the Radioactive Contamination due to Sr^{90} and Cs^{137})

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Theoretical Analysis of the Radioactive Contamination due to Sr^{90} and Cs^{137}

In view of the considerably different values on the fallout rate and the cumulative deposition of Sr^{90} and Cs^{137} reported from different parts of Japan, the author attempted to estimate the ranges of the fallout rate and of the cumulative deposition of Sr^{90} and Cs^{137} for different parts of Japan based upon the theoretical consideration.

(I) Theoretical Consideration[1][2]

About 10% of the deposited Sr^{90} has been reported to have come from tropospheric fallout in areas far from test sites, while about 30% may be taken as the representative value of the tropospheric fallout for areas relatively close to test sites.[2] Therefore, in areas far from test sites, it may be assumed that a major part of the radioactive fallout of the nuclides with relatively long half lives might be ascribed to the stratospheric fallout, although these percentages may be greatly different depending on the types of nuclear detonations. In case of the air burst, a greater part of the radioactivity may be injected into the stratosphere. In case of the ground surface explosion, a greater part of the radioactivity may be adsorbed or included in a large amount of dust particles with greatly different sizes and fall to the ground with the greatly different rate according to the sizes of the dust particles. In this case the fraction of local fallout may be higher as compared with the distant tropospheric fallout or stratospheric fallout. Since larger particles may fall faster than the smaller ones, the fraction of the dust falling to the ground per unit time may be considered to be a function of time after detonation at least during the initial period after the ground surface explosion. Therefore, in the tropospheric fallout

or especially in the local fallout at a relatively short time following the surface explosion there may be the case where the fraction deposited per unit time decreases with time because of the greatly different particle sizes of the dust which occluded or adsorbed the fission products and other induced activities. In this case if we assume that the fraction of the radioactive dust deposited per unit time decreases inversely proportional to the time t after explosion, the following expression may hold.

$$\frac{dD_a}{dt} \frac{1}{D_a} = -\frac{\beta}{t} \qquad (1)$$

where D_a is the amount of dust in the atmosphere and β a constant which may be different depending on the types and the conditions of nuclear detonation. Integrating the above equation, we may obtain,

$$D_a = D_1 t^{-\beta} \qquad (2)$$

where D_1 is a constant corresponding to the amount of dust in the atmosphere at unit time after detonation.

If we express the activity of the dust with I_a corresponding to D_a and if we assume that the grossactivity of the dust may decrease according to the Way-Wigner law $t^{-\alpha}$, the total activity remaining in the atmosphere may be expressed by the following power function of the time t after detonation.

$$I_a = I_1 t^{-(\alpha+\beta)} \qquad (3)$$

where I_1 is the activity in the atmosphere at unit time after detonation.

If we assume that the total amount of the radioactive dust particles of widely different sizes injected into the stratospheric reservoir would also decrease according to the following power function during the initial period,

$$D_s = D_1 t^{-\beta} \qquad (4)$$

the total amount of dust in the stratosphere D_t after τ days of continuous injection of an amount q per unit time may be given by the following equation.

$$D_t = R_1 q \int_o^\tau (\tau - t)^{-\beta} \, dt \qquad (5)$$

or, when β is smallr than unity

$$D_t = R_1 q \, \frac{\tau^{1-\beta}}{1-\beta} \qquad (6)$$

where R_1 is the fraction of the dust retained at end of unit time.

If we pay special attention to a certain specific radioactive nuclide which decays exponentially with the disintegration constant λ, the rate of change of the activity I_s of the specific nuclide corresponding to D_s in the stratosphere may be given by the following differential equation.

$$\frac{dI_s}{dt} = - \frac{\beta}{t} I_s - \lambda I_s \qquad (7)$$

Solving the above equation, we may obtain

$$I_s = I_1 \, t^{-\beta} \, e^{-\lambda(t-1)} \qquad (8)$$

where I_1 is the activity at t=1.

The total activity of the specific radioactive nuclide in the tratosphere I_t after τ days of continuous injection of the activity q_s of the specific nuclide per unit time may be given by the following equation.

$$I_t = R_1 \, q_s \int_o^\tau (\tau - t)^{-\beta} \, e^{-\lambda(\tau - t - 1)} \, dt \qquad (9)$$

where R_1 is the fractional retention at end of unit time.
From the definition of the incomplete gamma function,

$$\Gamma_z(n) = \int_0^z e^{-x} x^{n-1} dx \qquad (10)$$

Using the above relation, the equation (9) may be expressed as

$$I_{(t)} = R_1 q_{0s} e^{\lambda} \lambda^{\beta-1} \Gamma_{\lambda\tau}(1-\beta) \qquad (11)$$

However, sometime after the nuclear detonation we may assume that the size of the radioactive dust containing a certain specific nuclide in the stratospheric reservoir would be more or less uniform. In such cases, in view of the absence of the knowledge on the detailed mechanism of transport of the fine radioactive dust from the stratosphere to the troposphere it may be assumed as a working hypothesis that a constant fraction of the total amount in the stratosphere would be transported down to the troposphere per unit time. Under this assumption, if we assume that a major part of such long-lived fission products as Sr^{90} and Cs^{137} produced by high yield detonations comes from stratospheric fallout, the change of the stratospheric content of these nuclides with time may be expressed by the following differential equation.

$$\frac{dQ_s(t)}{dt} = n - \lambda Q_s(t) - F_r(t) \qquad (12)$$

where n is the injection rate of Sr^{90} or Cs^{137} into the stratosphere per unit area (mc/km^2 - year), $Q_{s(t)}$ the Sr^{90} or Cs^{137} content of the stratosphere expressed per unit area (mc/km^2), λ the disintegration constant of Sr^{90} or Cs^{137} (0.025/year), and $F_{r(t)}$ the average fall-out rate of Sr^{90} or Cs^{137} per unit area (mc/km^2 - year). For the calculation, $F_{r(t)}$ is assumed to be proportional to $Q_{s(t)}$ as a working hypothesis.

$$F_{r(t)} = kQ_{s(t)} \qquad (13)$$

where the elimination constant k may be expressed with the Half elimination time T_s by the following equation

$$k = \frac{0.693}{T_s} \qquad (14)$$

Under the above assumption, the general solution of equation (12), taking the relation (13) into consideration, may be expressed by

$$F_{r(t)} = F_{r(0)}\, e^{-(\lambda+k)t} + \frac{kn}{\lambda+k}\left[1 - e^{-(\lambda+k)t}\right] \qquad (15)$$

Using the above expression of fallout rate $F_r(t)$, the rate of the total cumulative deposition and the ground accumulation in the surface layer of the soil may be expressed by the following differential equations.

$$\frac{dF_{d(t)}}{dt} = F_{r(t)} - \lambda F_{d(t)} \qquad (16)$$

$$\frac{dF_{g(t)}}{dt} = F_{r(t)} - (\lambda+\ell)\, F_{g(t)} \qquad (17)$$

where $F_{d(t)}$ is the total cumulative deposition, $F_{g(t)}$ the ground accumulation in the surface layer of the soil, λ the disintegration constant of Sr^{90} or Cs^{137}, and ℓ the elimination constant indicating the annual fraction eliminated from the surface layer of the soil due to various weathering effects, which may greatly depend on the nature of the soil, amount of rainfall and other artificial factors such as irrigation, farming and ploughing. Using the half elimination time T_g due to weathering, the weathering constant l may be expressed by

$$\ell = \frac{0.693}{T_g} \qquad (18)$$

The general solutions of equations (16) and (17) may be given by the following equations.

$$F_{d(t)} = F_{d(0)}\, e^{-\lambda t} + \frac{F_{r(0)}}{k}\left[e^{-\lambda t} - e^{-(\lambda+k)t}\right] + \frac{n}{\lambda}\left[\frac{k}{\lambda+k} + \frac{\lambda}{\lambda+k}\, e^{-(\lambda+k)t} - e^{-\lambda t}\right] \qquad (19)$$

When $l \neq k$,

$$F_{g(t)} = F_{g(0)}\, e^{-(\lambda+\ell)t} + \frac{1}{k-\ell} F_{r(0)} \left[e^{-(\lambda+\ell)} - e^{-(\lambda+k)t} \right]$$

$$+ kn \left[\frac{1}{(\lambda+k)(\lambda+\ell)} - \frac{1}{(\lambda+\ )(k-\ell)} e^{-(\lambda+\ell)t} \right.$$

$$\left. + \frac{1}{(\lambda+k)(k-\ell)} e^{-(\lambda+k)t} \right] \qquad (20)$$

When $l=k$,

$$F_{g(t)} = F_{g(0)}\, e^{-(\lambda+k)t} + F_{r(0)} t e^{-(\lambda+k)t}$$

$$+ \frac{kn}{(\lambda+k)^2} \left[1 - \left\{ 1 + (\lambda+k)t \right\} e^{-(\lambda+k)t} \right] \qquad (21)$$

When we could assume $F_{d(0)} = 0$, $F_{g(0)} = 0$ and $F_{r(0)} = 0$, the above expressions of $F_{r(t)}$, $F_{d(t)}$ and $F_{g(t)}$ may be written as follows:

$$F_{r(t)} = \frac{kn}{\lambda+k} \left[1 - e^{-(\lambda+k)t} \right] \qquad (22)$$

$$F_{d(t)} = \frac{n}{\lambda} \left[\frac{k}{\lambda+k} + \frac{\lambda}{\lambda+k} e^{-(\lambda+k)t} - e^{-\lambda t} \right] \qquad (23)$$

When $l \neq k$,

$$F_{g(t)} = kn \left[\frac{1}{(\lambda+k)(\lambda+\ell)} - \frac{1}{(\lambda+\ell)(k-\ell)} e^{-(\lambda+\ell)t} \right.$$

$$\left. + \frac{1}{(\lambda+k)(k-\ell)} e^{-(\lambda+k)t} \right] \qquad (24)$$

When $l=k$,

$$F_{g(t)} = \frac{kn}{(\lambda+k)^2} \left[1 - \left\{ 1 + (\lambda+k)t \right\} e^{-(\lambda+k)t} \right] \qquad (25)$$

The equations (22) - (25) may be applicable approximately in the analysis of the fallout effects during the period from March 1954 to the end of 1958 in the areas where $F_{d(0)}$ and $F_{r(0)}$ in March 1954 could be considered negligible.

After the tests were stopped towards the end of 1958, n=0 until the resumption of tests which would significantly contribute to the stratospheric reservoir of Sr^{90} or Cs^{137}. Therefore, substituting n=0 into the equations (15)and (19) - (21) and assuming t=0 at the end of 1958, we may obtain the following expressions for the period after the end of 1958.

$$F_{r(t)} = F_{r(0)}\, e^{-(\lambda+k)t} \tag{26}$$

$$F_{d(t)} = F_{d(0)} e^{-\lambda t} + \frac{F_{r(0)}}{k}\left[e^{-\lambda t} - e^{-(\lambda+k)t}\right] \tag{27}$$

When l≠k,

$$F_{g(t)} = F_{d(0)} e^{-(\lambda+l)t} + \frac{1}{k-l} F_{r(0)}\left[e^{-(\lambda+l)t} - e^{-(\lambda+k)t}\right] \tag{28}$$

When l=k,

$$F_{g(t)} = F_{g(0)} e^{-(\lambda+k)t} + F_{r(0)} t e^{-(\lambda+k)t} \tag{29}$$

The above equations indicate that the fallout rate $F_{r(t)}$ decreases after the suspension of the tests, but the total fallout cumulative deposition $F_{d(t)}$ and the fallout ground accumulation $F_{g(t)}$ may be expected to increase for some time and then begin to decrease after going through a maximum point. The time, at which the maximum point will be reached in $F_{d(t)}$, may be obtained, by solving the equation $\frac{dF_{d(t)}}{dt} = 0$, to be

$$t_{max} = \frac{1}{k} \ln \frac{(\lambda+k) F_{r(0)}}{\lambda[k F_{d(0)} + F_{r(0)}]} \tag{30}$$

Similarly, the time corresponding to the maximum point in $F_{g(t)}$ may be obtained, by solving the equation $\frac{dF_{g(t)}}{dt} = 0$, to be as follows:

When $l \neq k$,

$$t_{max} = \frac{1}{k-l} \ln \frac{(\lambda+k) F_{r(0)}}{(\lambda+l)[(k-l) F_{g(0)} + F_{r(0)}]} \tag{31}$$

When $l=k$,

$$t_{max} = \frac{F_{r(0)} - (\lambda+k) F_{g(0)}}{(\lambda+k) F_{r(0)}} \tag{32}$$

If we assume that the nuclear tests should continue indefinitely with the constant value of n, the fallout rate in equation (15) may be expected to reach the following equilibrium value.

$$F_{r}(t \to \infty) = \frac{kn}{\lambda+k} \tag{33}$$

Similary, the total deposition $F_{d(t)}$ of equation (19) and the ground accumulation in the surface layer $F_{g(t)}$ of equation (20) may be expected to reach the following equilibrium values.

$$F_{d}(t \to \infty) = \frac{kn}{\lambda(\lambda+k)} \tag{34}$$

$$F_{g}(t \to \infty) = \frac{kn}{(\lambda+l)(\lambda+k)} \tag{35}$$

However, since the tests were suspended towards the end of 1958, the average injection rate $\bar{n}$ for the period 1954 to 1958 may be estimated from the following relation.

$$\bar{n} = \frac{\lambda}{(1-e^{-\lambda t})} \left[F_{d(t)} + \frac{1}{k} F_{r(t)} \right] \tag{36}$$

where t may be taken to be 5 years, assuming t=0 early in 1954.

(II) Results of Theoretical Analysis

Assuming the cumulative deposition $F_{d(t)}$ of Sr^{90} to be about 10 - 15 mc/km^2 and the fallout rate $F_{r(t)}$ of Sr^{90} to be about 3 - 5 mc/km^2 - year at the end of 1958 and the half elimination time Ts from the stratospheric reservoir to be about 2 years, the average value of the annual injection rate during the period from early 1954 to the end of 1958 may be estimated from the equation (36) to be approximately $\bar{n}$ = 4.3 - 6.3 mc/km^2 - year. Assuming the uniform distribution of Sr^{90} over the entire surface of the earth, the specific concentration of Sr^{90} per unit area may be estimated at about 3.6 mc/km^2 corresponding to the fast neutron fission of U^{238} equivalent to about 20 megatons of TNT.(3) Comparing with this value, the above value of $\bar{n}$ may be considered to correspond to the annual injection rate of about 24 - 35 megatons TNT equivalent fission products, or about 120 - 175 megatons fission yield. If we assume that the above value of $\bar{n}$ as estimated from the data observed in the Pacific coast of Japan would be about three times higher than the world average and that roughly about one half of the total fission products entered the stratospheric reseroir on an average, the total fission yield during the five year period from early 1954 to the end of 1958 may be estimated to be roughly about 90 - 120 megatons equivalent.

Fig.1 shows the range of fallout rate of Sr^{90} during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (22), assuming $F_{r(0)}$=0 in equation (15) early 1954 and n=4.3 and 6.3 mc/km^2-year, and the half deposition time T_s of about 2 years. In the figure diffarent estimates of the fallout rate of Sr^{90} for Osaka and Tokyo based upon the observations at different institutes are also shown. In this figure, Osaka I indicates the data reported by the Japan

←(Fig.1)

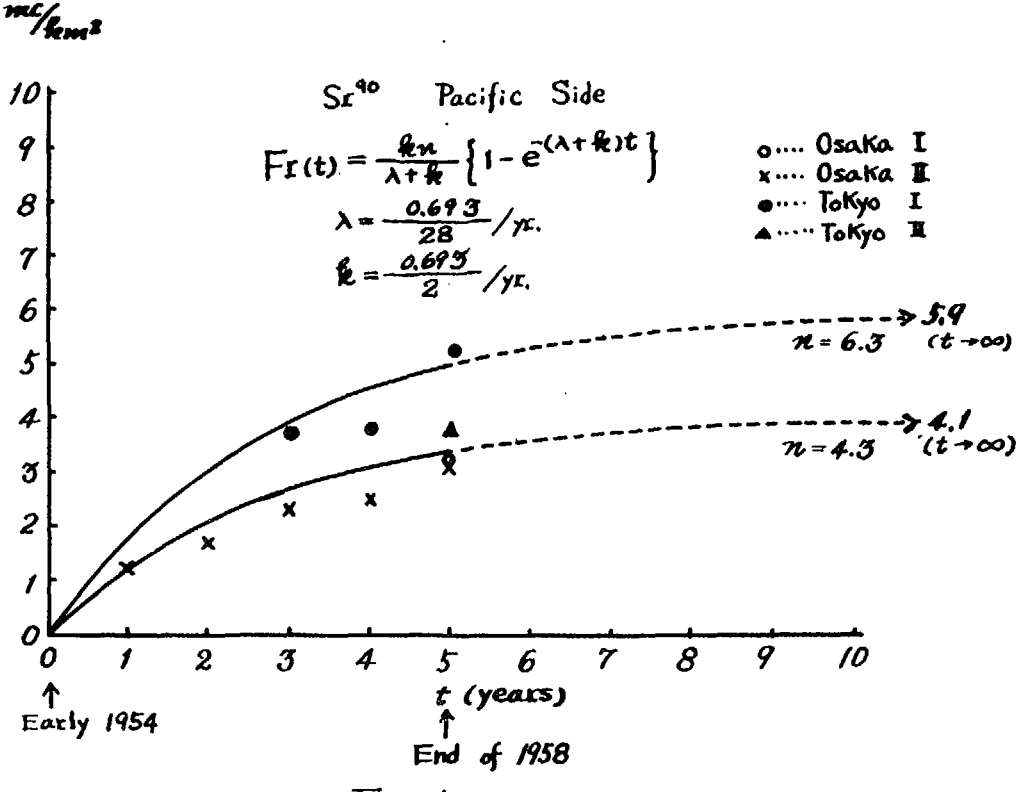

mc/km²
Sr⁹⁰ Pacific Side
Fr(t) = kn/(λ+k) {1 − e^−(λ+k)t}
λ = 0.693/28 /yr.
k = 0.693/2 /yr.
o.... Osaka I
x.... Osaka II
●.... Tokyo I
▲.... Tokyo II
5.9
n = 6.3 (t→∞)
4.1
n = 4.3 (t→∞)
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 1

Meteorological Agency in Tokyo for Osaka,[4] Osaka II those by the author,[5] and Tokyo I is a combination of the data given in the " Survey of Radioactivity " compiled by the Japan Science and Technology Agency[6] and those given by the Japan Meteorological Agency,[4] and Tokyo II is the data given by Izawa, et al[7]. As can be seen in the figure the range of fallout rate of Sr^{90} at the Pacific side of the Japan mainland may be estimated to be about 3 - 5 mc/km^2 - year at the end of 1958. However, if the above rates of injection should continue indefinitely, the fallout rate of Sr^{90} may be expected to reach the equilibrium value of about 4.1 - 5.9 mc/km^2 - year as estimated from the equation (33).

Fig.2 shows the range of fallout rate of Cs^{137} during the period from April 1954 to the end of August 1958 at the Pacific side of the Japan mainland as estimated from the equation (22), assuming n=8.3 - 18.9 mc/km^2 - year. In this case, since the ratio Cs^{137}/Sr^{90} may be estimated to be about 1.93[3] from the fission yield curve for U^{238}, the lower limit of n for Cs^{137} is assumed to be 1.93 times higher than the lower limit n=4.3 for Sr^{90}. The upper limit of n for Cs^{137} is assumed to be about 3 times that of Sr^{90}. ←(Fig.2)

In this figure, Osaka I is the data given by the author,[5] and Osaka II is the value estimated by multiplying the value for Sr^{90} by a factor of 2.8 which is the ratio Cs^{137}/Sr^{90} estimated for Tokyo.[7] Tokyo I is the data given by the Japan Meteorological Agency in Tokyo[4] and Tokyo II those given by Izawa, et al.[7] As can be seen in the figure, the range of the fallout rate of Cs^{137} at the Pacific side of the Japan mainland may be estimated to be about 6.5 - 14.8 mc/km^2 - year at the end of 1958.

However if the above injection rate should continue indefinitely, the fallout rate of Cs^{137} may be expected to reach the equilibrium value of about 7.7 - 17.6 mc/km^2 - year.

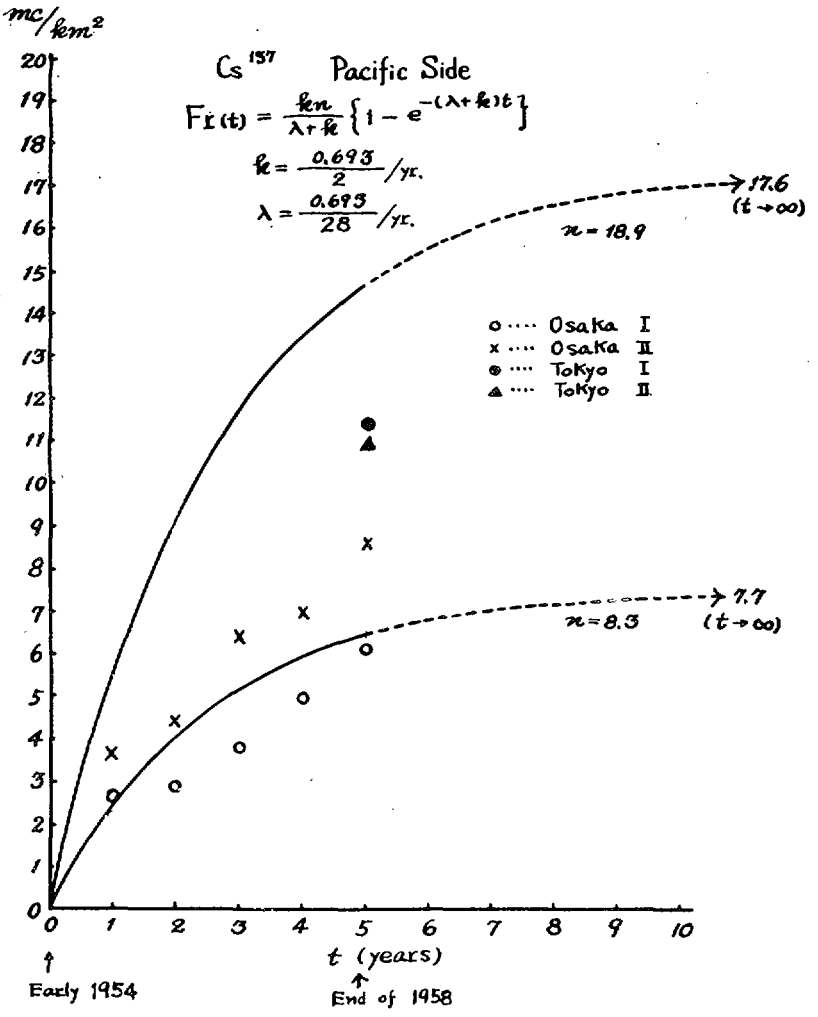

mc/km²
Cs 137 Pacific Side
Fr(t) = kn/(λ+k) {1 − e^−(λ+k)t}
k = 0.693/2 /yr.
λ = 0.693/28 /yr.
17.6 (t→∞)
n = 18.9
o ···· Osaka I
x ···· Osaka II
● ···· Tokyo I
▲ ···· Tokyo II
7.7 (t→∞)
n = 8.3
t (years)
Early 1954
End of 1958

Fig. 2

Fig.3 shows the range of the fallout rate of Sr^{90} during the period from April 1954 to the end of August 1958 at the Japan Sea side of the Japan mainland as estimated from the equation (22), assuming n=12.6 - 18.9 mc/km^2 - year. The value of n was estimated by assuming two to three times higher at the Japan Sea side than the upper limit for the Pacific side. As can be seen in the figure, the range of fallout rate of Sr^{90} at the Japan Sea side of the Japan mainland may be estimated to be about 10 - 15 mc/km^2 - year at the end of 1958. However if the above injection rate should continue indefinitely, the fallout rate of Sr^{90} may be expected to reach the equilibrium value of about 12.6 - 18.9 mc/km^2 - year. ←(Fig.3)

Fig.4 shows the range of the fallout rate of Cs^{137} during the period from April 1954 to the end of 1958 at the Japan Sea side of the Japan mainland as estimated from the equation (22), assuming n=24.9 - 50.7 mc/km^2 - year. As can be seen in the figure, the range of fallout rate of Cs^{137} at the Japan Sea side may be estimated to be about 20 - 40 mc/km^2 - year. However, if the above injection rate should continue indefinitely, the fallout rate of Cs^{137} may be expected to reach the equilibrium value of about 23.2 - 47.2 mc/km^2 - year. ←(Fig.4)

Fig.5 shows the range of the cumulative deposition of Sr^{90} during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (19) assuming $F_{r(0)}$=0 and $F_{d(0)}$=0.5 mc/km^2 early 1954 and the same injection rate n=4.3 - 6.3 mc/km^2 - year as assumed at the estimation of the fallout rate. As can be seen in the figure, the range of cumulative deposition of Sr^{90} at the Pacific side of the Japan mainland may be estimated to be about 11 - 16 mc/km^2 at the end of 1958. ←(Fig.5)

In the figure different estimates of the cumulative deposition

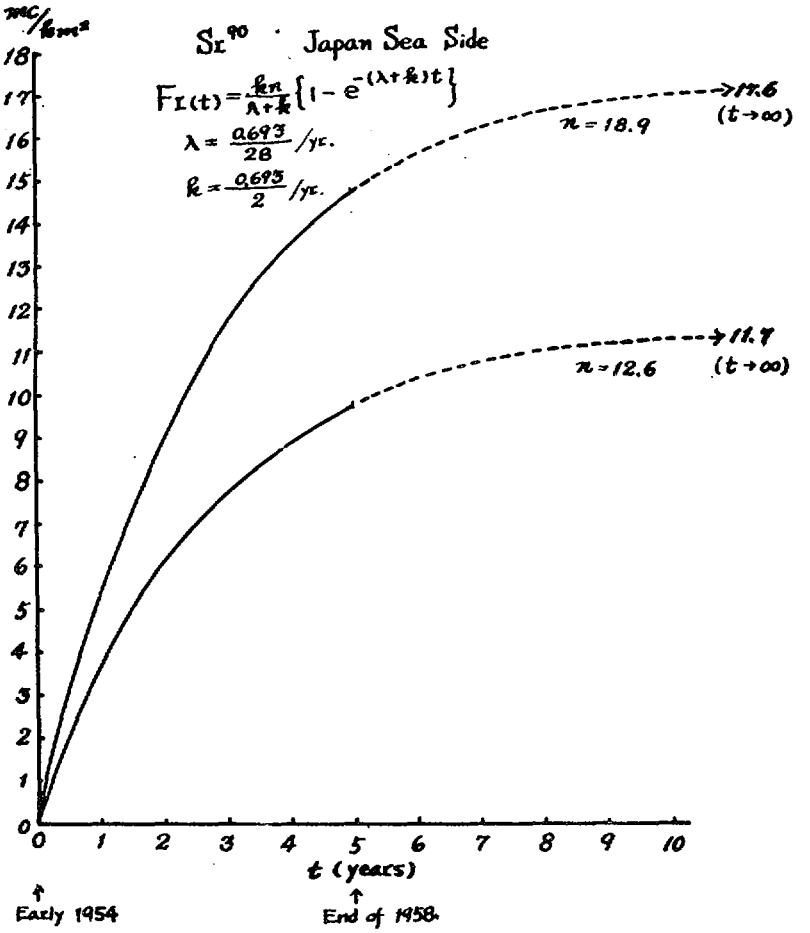

mc/km²
Sr⁹⁰ · Japan Sea Side
$F_I(t)=\frac{fn}{\lambda+k}\{1-e^{-(\lambda+k)t}\}$
$\lambda=\frac{0.693}{28}$/yr.
$k=\frac{0.693}{2}$/yr.
n = 18.9
→17.6 (t→∞)
n = 12.6
→11.7 (t→∞)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 3

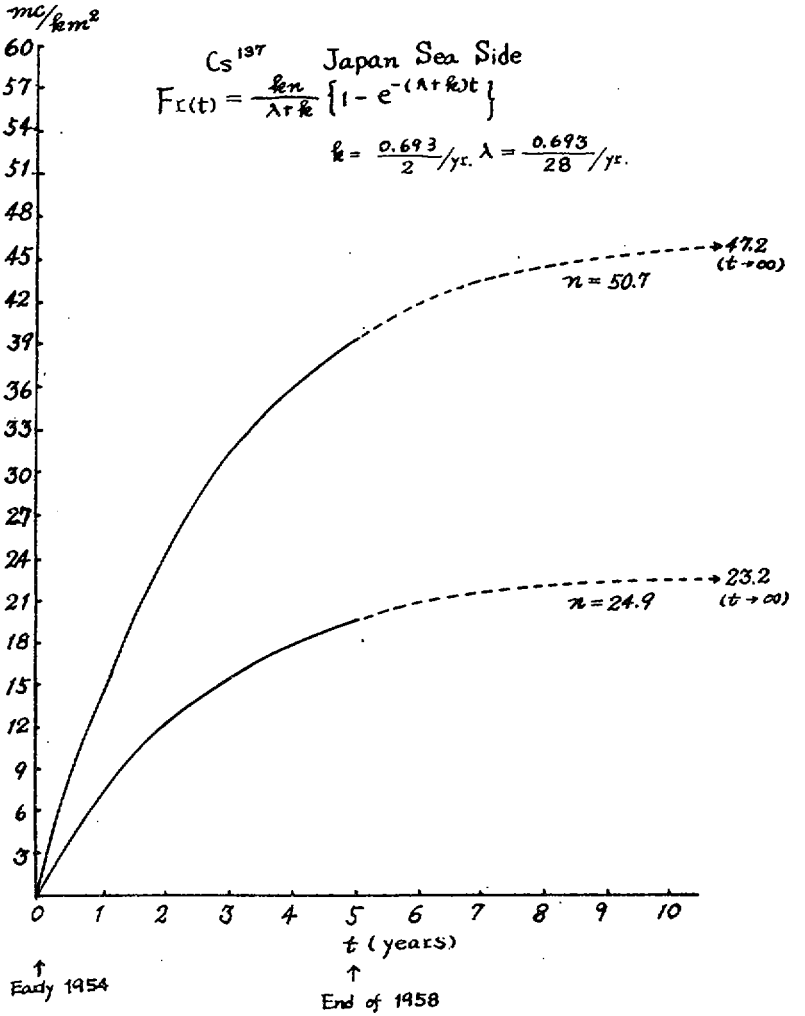

mc/km²
60
57
54
51
48
45
42
39
36
33
30
27
24
21
18
15
12
9
6
3
Cs¹³⁷ Japan Sea Side
$F_x(t) = \frac{kn}{\lambda + k}\{1 - e^{-(\lambda+k)t}\}$
$k = \frac{0.693}{2}$/yr. $\lambda = \frac{0.693}{28}$/yr.
47.2 (t→∞)
n = 50.7
23.2 (t→∞)
n = 24.9
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 4

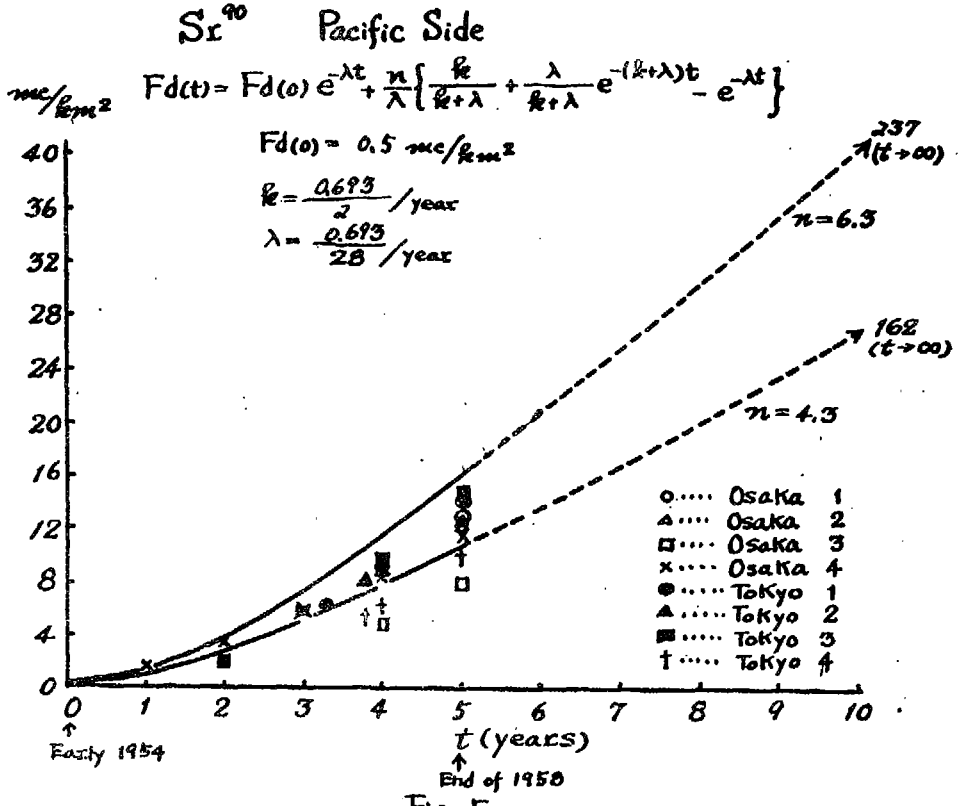

Sr90 Pacific Side
mc/km²
Fd(t) = Fd(0) e^{-λt} + n/λ { k/(k+λ) + λ/(k+λ) e^{-(k+λ)t} - e^{-λt} }
Fd(0) = 0.5 mc/km²
k = 0.693/2 /year
λ = 0.693/28 /year
237 (t→∞)
n=6.3
162 (t→∞)
n=4.3
Osaka 1
Osaka 2
Osaka 3
Osaka 4
Tokyo 1
Tokyo 2
Tokyo 3
Tokyo 4
t (years)
Early 1954
End of 1958

Fig. 5

of Sr^{90} for Osaka and Tokyo based upon the observations at different institutes are also shown. In this figure Osaka 1 indicates a combination of the data given for the cumulative deposition in Tokyo up to the end of 1957 in the " Survey of Radioactivity " compiled by the Japan Science and Technology Agency[6] plus the data given for Osaka by the Japan Meteorological Agency in Tokyo[4] after the end of 1957. In other words, since no data for Osaka prior to the end of 1957 had been given by the Japan Meteorological Agency, the cumulative deposition of Sr^{90} in Osaka was assumed to be the same as that in Tokyo at the end of 1957, to which the values given later for Osaka by the Japan Meteorological Agency[4] was added. Osaka 2 is a combination of the data given for the cumulative deposition in Tokyo at the end of 1957 by the Japan Meteorological Agency[4] plus those given for Osaka after the end of 1957 by the Japan Meteorological Agency in Tokyo.[4] However, according to the measurements by the Japan Meteorological Agency[4] for Osaka and Tokyo during the period from Jan. 1958 to June 1960, the data for Osaka is lower on an average by a factor 7/13.75. Therefore in Osaka 3 the cumulative deposition in Osaka at the end of 1957 was assumed to be lower by this factor than that estimated for Tokyo, to which the data given thereafter for Osaka by the Japan Meteorological Agency[4] was added.

Osaka 4 is the data given by the author.[5] Tokyo 1 is the data given for Tokyo by the Japan Meteorological Agency.[4] Tokyo 2 is a combination of the data given for the cumulative deposition in Tokyo at the end of July 1957 by the Japan Meteorological Agency[4] plus the data given for Tokyo by Izawa, et al[7] since Aug. 1957. However the overall ratio of the cumulative deposition of Sr^{90} estimated for Tokyo during the period from Aug.1957 to the end of July 1960 by the Japan Meteorological Agency[4] to that by Izawa, et al[7] is about 10.94/16.24. Therefore, in Tokyo 4 the cumulative deposition of Sr^{90} at the end of July 1957 in Tokyo estimated by the Japan Meteorological Agency[4] is reduced by the above

factor, to which the data given by Izawa, et al[7] for the period thereafter was added. As can be seen in the figure, if the above injection rate should continue indefinitely, the cumulative deposition of Sr^{90} may be expected to reach the equilibrium value of about 162 - 237 mc/km^2, as estimated from the equation (34).

Fig.6 shows the range of the cumulative depostion of Cs^{137} during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (19) assuming $F_{r(0)}=0$ and $F_{d(0)}=0.5$ mc/km^2 early 1954 and the injection rate n=8.3 - 18.9 mc/km^2 - year. In this figure, different estimates on the cumulative deposition of Cs^{137} for Osaka and Tokyo are also shown. Osaka 1 is the data given by the author[5] and Osaka 2 is the data estimated by multiplying the data of Sr^{90} given by the author by a factor of 2.8 which is the ratio Cs^{137}/Sr^{90} estimated for Tokyo.[7] Tokyo 1 is the data given by the Japan Meteorological Agency in Tokyo. Tokyo 2 is a combination of the data given for the cumulative deposition of Cs^{137} at the end of July 1957 in Tokyo by the Japan Meteorological Agency[4] plus the data given thereafter by Izawa, et al[7] for Tokyo.

However, the overall ratio of the cumulative deposition of Cs^{137} during the period from Aug. 1957 to the end of July 1960 in Tokyo estimated by Izawa, et al. to that by the Japan Meteorological Agency is about 30.87/43.8. Therefore, in Tokyo 3 the cumulative deposition of Cs^{137} at the end of July 1957 was reduced by the above factor, to which the data given thereafter by Izawa, et al was added. As can be seen in the figure, the range of the cumulative deposition of Cs^{137} at the Pacific side of the Japan mainland may be estimated to be about 20 - 46 mc/km^2 at the end of 1958. However, if the above injection rate should continue indefinitely, the cumulative deposition of Cs^{137} may be expected to reach the equilibrium value of about 475 - 714 mc/km^2.

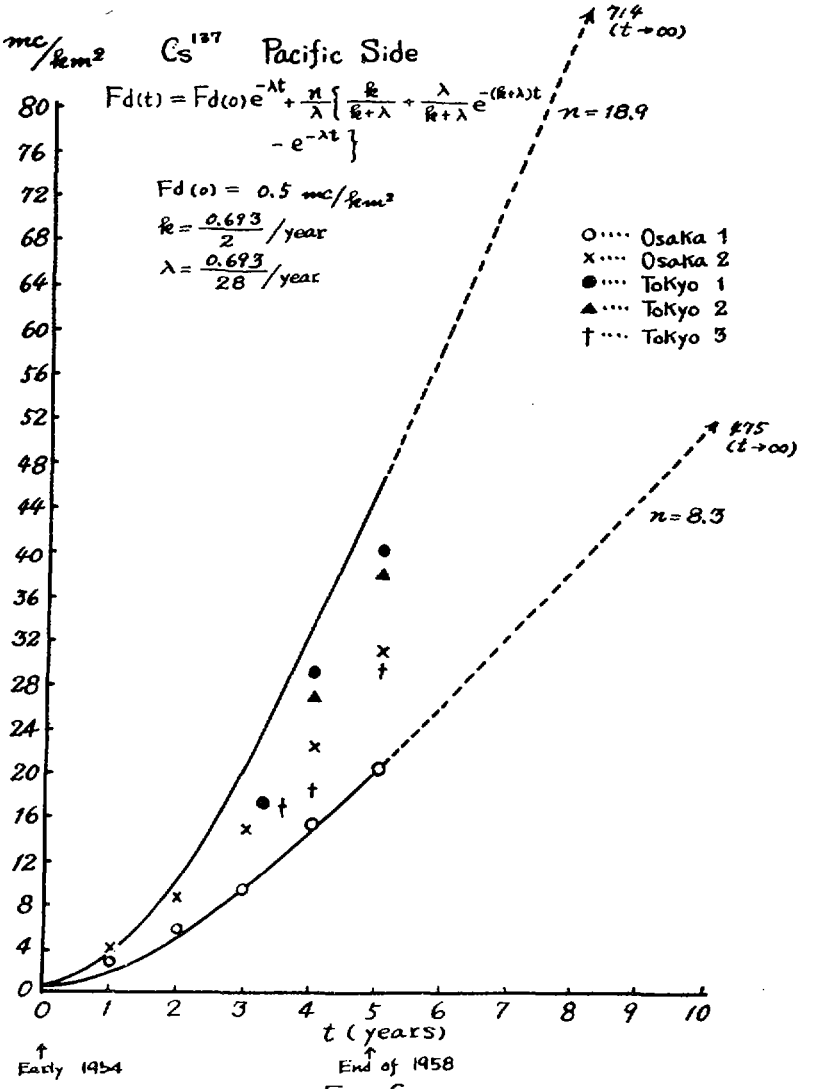

mc/km²
Cs¹³⁷ Pacific Side
Fd(t) = Fd(0)e^{-λt} + n/λ { k/(k+λ) + λ/(k+λ) e^{-(k+λ)t} - e^{-λt} }
Fd(0) = 0.5 mc/km²
k = 0.693/2 /year
λ = 0.693/28 /year
714 (t→∞)
n = 18.9
475 (t→∞)
n = 8.3
○···· Osaka 1
×···· Osaka 2
●···· Tokyo 1
▲···· Tokyo 2
†···· Tokyo 3
t (years)
Early 1954
End of 1958

Fig. 6

Fig.7 shows the range of the cumulative deposition of Sr^{90} during the period from April 1954 to the end of 1958 at the Japan Sea side of the Japan mainland as estimated from the equation (19) assuming $F_{r(0)}=0$ and $F_{d(0)}=0.5$ mc/km^2 early 1954 and the same injection rate n=12.6 - 18.9 mc/km^2 - year as assumed at the estimation of the fallout rate of Sr^{90} at the Japan Sea side. As can be seen in the figure, the range of the cumulative deposition of Sr^{90} at the Japan Sea side may be estimated to be about 32 - 46 mc/km^2 at the end of 1958. However, if the above injection rate should continue indefinitely, the cumulative deposition of Sr^{90} may be expected to reach the equilibrium value of about 475 - 714 mc/km^2. ←(Fig.7)

Fig.8 shows the range of the cumulative deposition of Cs^{137} during the period from April 1954 to the end of 1958 at the Japan Sea side of the Japan mainland as estimated from the equation (19) assuming $F_{r(0)}=0$ and $F_{d(0)}=0.5$ mc/km^2 early 1954 and the same injection rate n=24.9 - 50.7 mc/km^2 - year as assumed at the estimation of the fallout rate of Cs^{137} at the Japan Sea side. As can be seen in the figure, the range of the cumulative deposition of Cs^{137} at the Japan Sea side may be estimated to about 55 - 125 mc/km^2. ←(Fig.8)

However, if the above injection rate should continue indefinitely, the cumulative deposition of Cs^{137} may be expected to reach the equilibrium level of about 940 - 1,920 mc/km^2.

Fig.9 shows the range of the ground accumulation of Sr^{90} in the surface layer of the soil during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (20) assuming $F_{r(0)}=0$ and $F_{g(0)}=0.5$ mc/km^2 and the half elimination time T_e due to weathering to be about 10 years and n=4.3 - 6.3 mc/km^2 - year. In the figure, the ground accumulation ←(Fig.9)

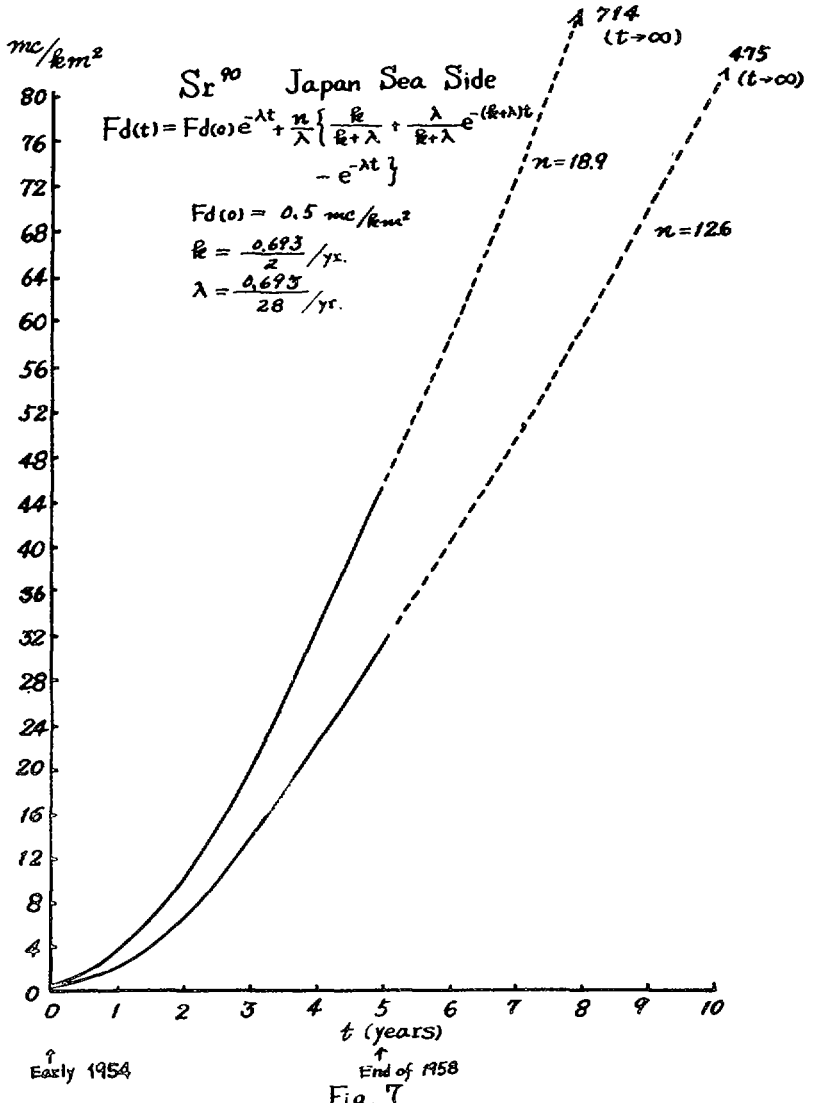

mc/km²
Sr⁹⁰ Japan Sea Side
Fd(t) = Fd(0) e^{-λt} + n/λ { k/(k+λ) + λ/(k+λ) e^{-(k+λ)t} - e^{-λt} }
Fd(0) = 0.5 mc/km²
k = 0.693/2 /yr.
λ = 0.693/28 /yr.
714 (t→∞)
475 (t→∞)
n=18.9
n=12.6
80
76
72
68
64
60
56
52
48
44
40
36
32
28
24
20
16
12
8
4
0
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 7

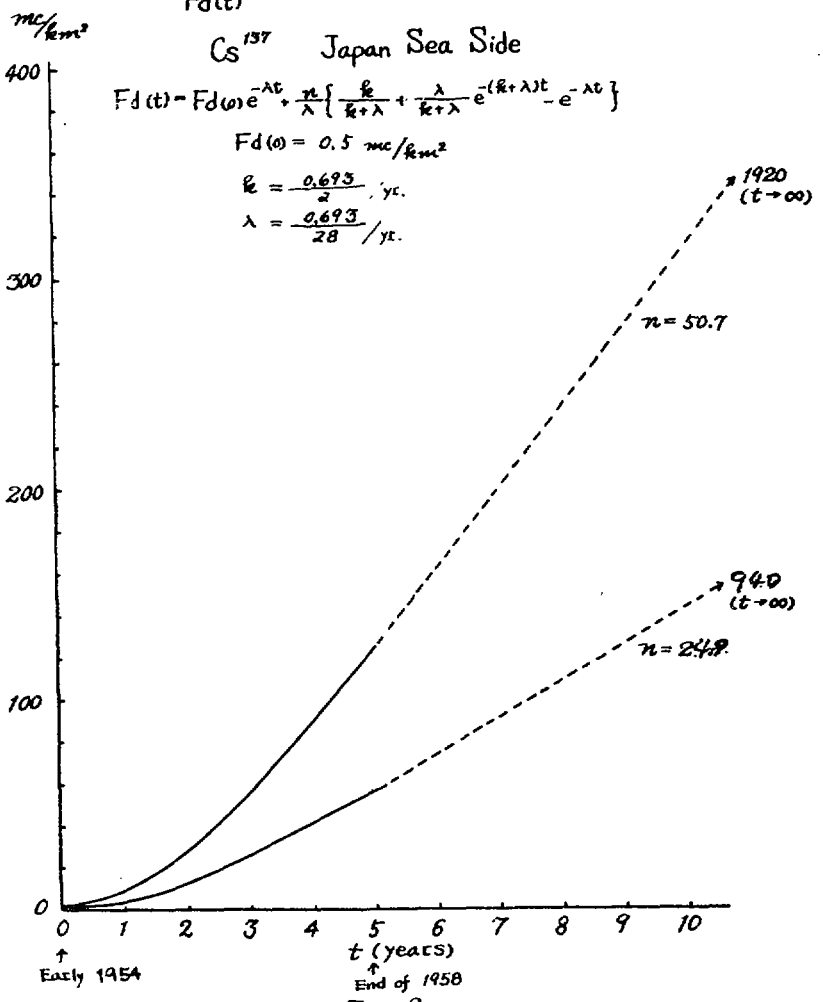

Fd(t)
mc/km²
Cs¹³⁷ Japan Sea Side
Fd(t) = Fd(0)e^{-λt} + n/λ { k/(k+λ) + λ/(k+λ) e^{-(k+λ)t} - e^{-λt} }
Fd(0) = 0.5 mc/km²
k = 0.693/2 /yr.
λ = 0.693/28 /yr.
1920 (t→∞)
n = 50.7
94.0 (t→∞)
n = 24.8
400
300
200
100
0
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 8

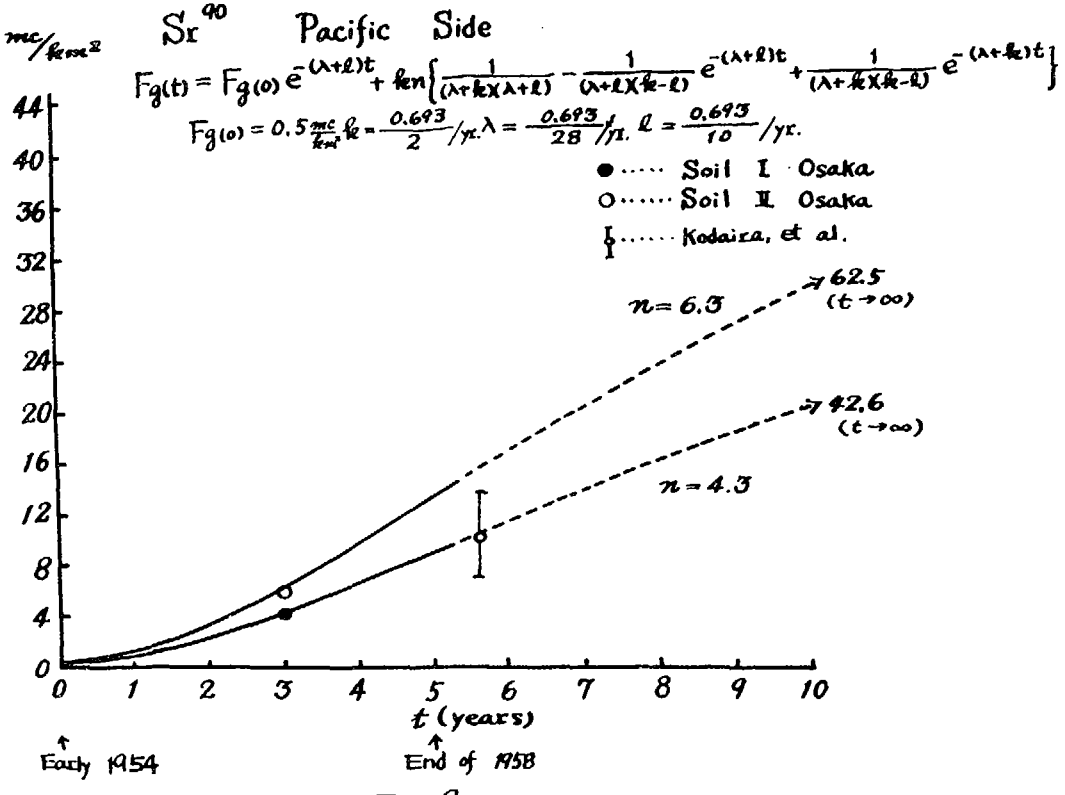

Sr 90 Pacific Side
mc/km²
$F_{g(t)} = F_{g(0)} e^{-(\lambda+\ell)t} + kn\left[\frac{1}{(\lambda+k)(\lambda+\ell)} - \frac{1}{(\lambda+\ell)(k-\ell)} e^{-(\lambda+\ell)t} + \frac{1}{(\lambda+k)(k-\ell)} e^{-(\lambda+k)t}\right]$
$F_{g(0)} = 0.5 \frac{mc}{km^2} k = \frac{0.693}{2}/yr. \lambda = \frac{0.693}{28}/yr. \ell = \frac{0.693}{10}/yr.$
● ····· Soil I Osaka
○ ····· Soil II Osaka
I ····· Kodaira, et al.
n = 6.3
62.5 (t→∞)
n = 4.3
42.6 (t→∞)
44
40
36
32
28
24
20
16
12
8
4
0
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 9

of Sr^{90} in the surface layer of soil as estimated by the author based upon the radiochemical analysis of the soil conducted early 1957 in Osaka[5] and the range of the ground accumulation of the exchangeable Sr^{90} in the agricultural soil estimated by Kodaira, et al[8] for the Pacific side of the Japan mainland based upon the radiochemical analysis of the soil sampled in June 1959 are also shown. As can be seen in the figure, the ground accumulation of Sr^{90} in the surface layer of the soil at the Pacific side of the Japan mainland may be estimated to be about 9.5 - 13.8 mc/km^2 at the end of 1958.

However, if the above injection rate should continue indefinitely, the ground accumulation of Sr^{90} in the surface layer of soil may be expected to reach the equilibrium value of about 42.6 - 62.5 mc/km^2, as estimated from the equation (35).

Fig.10 shows the range of the ground accumulation of Cs^{137} in the surface layer of the soil during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (20) assuming $F_{r(0)}=0$ and $F_{g(0)}=0.5$ mc/km^2 and the half elimination time T_g due to weathering to be about 10 years and n=8.3 - 18.9 mc/km^2 - year. In the figure, the ground accumulation of Cs^{137} in the surface layer of soil as estimated by multiplying the range of Sr^{90} at the Pacific side in June 1959 by a factor of 2.8 is also shown. ←(Fig.10)

As can be seen in the figure, the ground accumulation of Cs^{137} in the surface layer of the soil at the Pacific side of the Japan mainland may be estimated to be about 18 - 41 mc/km^2 at the end of 1958. However, if the above injection rate should continue indefinitely, the ground accumulation of Cs^{137} in the surface layer of soil may be expected to reach the equilibrium value of about 82.4 - 187.5 mc/km^2.

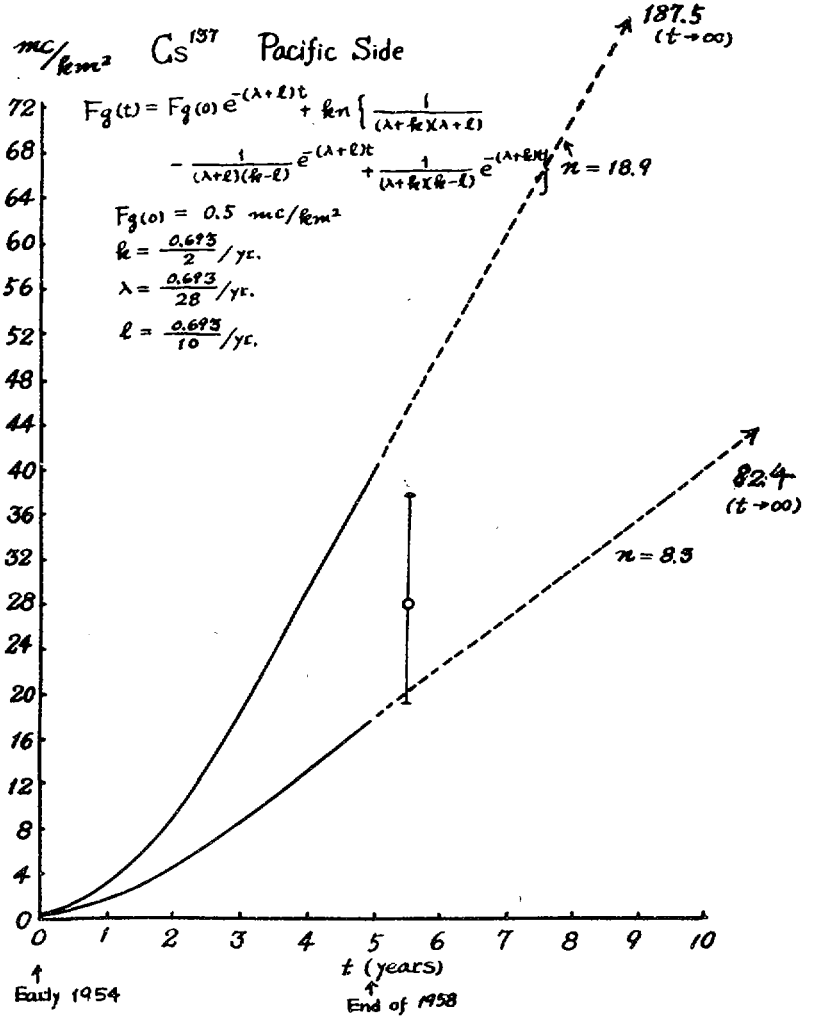

mc/km² Cs¹³⁷ Pacific Side
$F_g(t) = F_g(0)\, e^{-(\lambda+\ell)t} + kn\left\{\frac{1}{(\lambda+k)(\lambda+\ell)} - \frac{1}{(\lambda+\ell)(k-\ell)} e^{-(\lambda+\ell)t} + \frac{1}{(\lambda+k)(k-\ell)} e^{-(\lambda+k)t}\right\}$
$F_g(0) = 0.5$ mc/km²
$k = \frac{0.693}{2}$ /yr.
$\lambda = \frac{0.693}{28}$ /yr.
$\ell = \frac{0.693}{10}$ /yr.
187.5 (t→∞)
n = 18.9
82.4 (t→∞)
n = 8.3
72 68 64 60 56 52 48 44 40 36 32 28 24 20 16 12 8 4 0
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 10

Fig.11 shows the range of the ground accumulation of Sr^{90} in the surface layer of the soil during the period from April 1954 to the end of 1958 at the Pacific side of the Japan mainland as estimated from the equation (20) assuming $F_{r(0)}=0$ and $F_{g(0)}=0.5$ mc/km^2 and the half elimination time T_g due to weathring to be about 10 years and n=12.6 - 18.9 mc/km^2 - year. In the figure, the ground accumulation of the exchangeable Sr^{90} in the surface layer of agricultural soil estimated by Kodaira, et al[8] for the Japan Sea side of the Japan mainland based upon the radiochemical analysis of the soil sampled in June 1959 are also shown. ←(Fig.11)

As can be seen in the figure, the ground accumulation of Sr^{90} in the surface layer of the soil at the Japan Sea side of the Japan mainland may be estimated to be about 27 - 41 mc/km^2 at the end of 1958.

However, if the above rate of injection should continue indefinitely, the ground accumulation of Sr^{90} in the surface layer of the soil may be expected to reach the equilibrium value of about 125 - 187.5 mc/km^2.

Fig.12 shows the range of the ground accumulation of Cs^{137} in the surface layer of the soil during the period from April 1954 to the end of 1958 at the Japan Sea side of the Japan mainland as estimated from the equation (20) assuming $F_{r(0)}=0$ and $F_{g(0)}=0.5$ mc/km^2 and the half elimination time T_g due to weathering to be about 10 years and n=24.9 - 50.7 mc/km^2 - year. In the figure, the ground accumulation of Cs^{137} in the surface layer of the soil as estimated by multiplying that of Sr^{90} at the Japan Sea side in June 1959 by a factor of 2.8 is also shown. ←(Fig.12)

As can be seen in the figure, the ground accumulation of Cs^{137} in the surface layer of the soil at the Japan Sea side of the Japan mainland may be estimated to be about 53 - 108 mc/km^2 at the end of 1954.

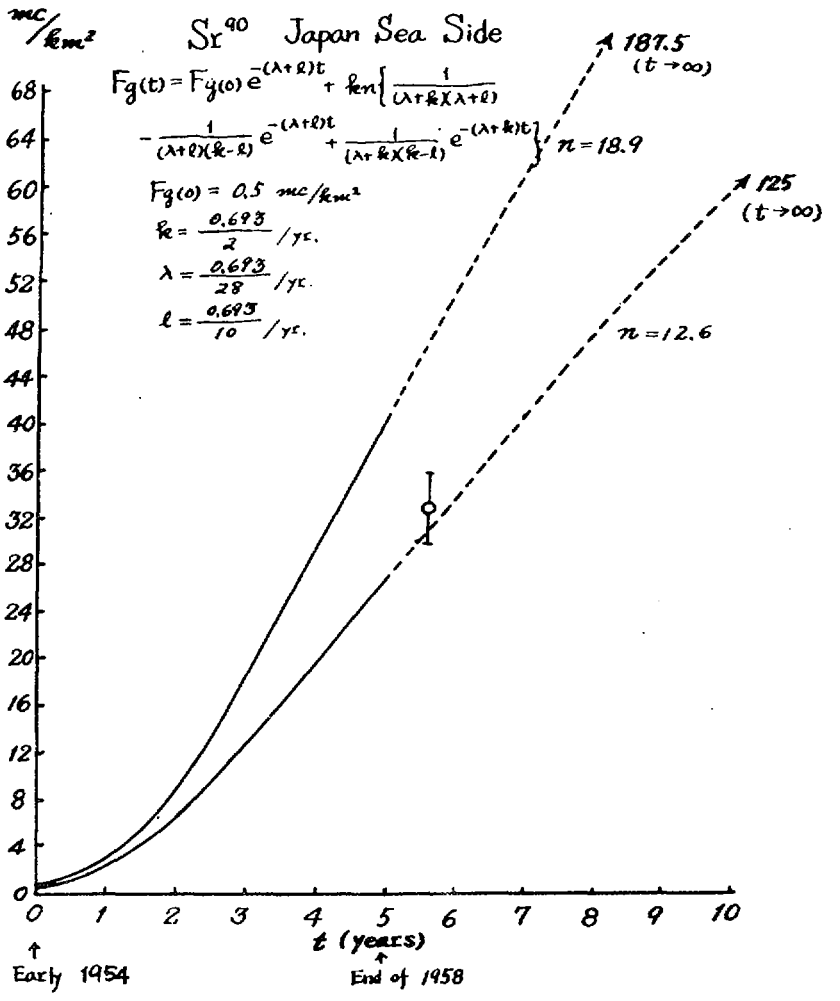
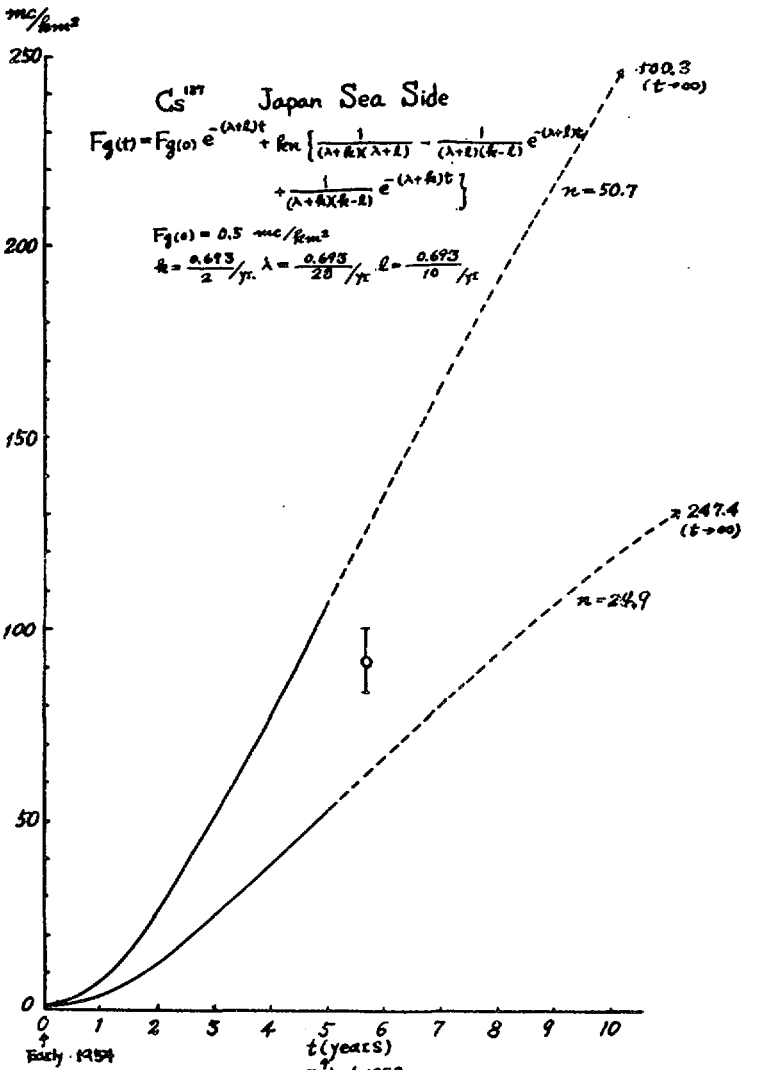

mc/km²
Sr⁹⁰ Japan Sea Side
Fg(t) = Fg(0) e^{-(λ+ℓ)t} + kn{ 1/((λ+k)(λ+ℓ)) - 1/((λ+ℓ)(k-ℓ)) e^{-(λ+ℓ)t} + 1/((λ+k)(k-ℓ)) e^{-(λ+k)t} }
Fg(0) = 0.5 mc/km²
k = 0.693/2 /yr.
λ = 0.693/28 /yr.
ℓ = 0.693/10 /yr.
187.5 (t→∞)
n = 18.9
125 (t→∞)
n = 12.6
68
64
60
56
52
48
44
40
36
32
28
24
20
16
12
8
4
0
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958

Fig. 11


mc/km²
250
200
150
100
50
0
Cs¹³⁷ Japan Sea Side
$F_g(t) = F_{g(0)} e^{-(\lambda+\ell)t} + kn\left\{\frac{1}{(\lambda+k)(\lambda+\ell)} - \frac{1}{(\lambda+\ell)(k-\ell)} e^{-(\lambda+k)t} + \frac{1}{(\lambda+k)(k-\ell)} e^{-(\lambda+k)t}\right\}$
$F_{g(0)} = 0.5$ mc/km²
$k = \frac{0.693}{2}$/yr. $\lambda = \frac{0.693}{28}$/yr $\ell = \frac{0.693}{10}$/yr
x 100.3 (t→∞)
n = 50.7
x 247.4 (t→∞)
n = 24.9
0 1 2 3 4 5 6 7 8 9 10
t (years)
Early 1954
End of 1958
Fig. 12

However, if the above injection rate should continue indefinitely, the ground accumulation of Cs^{137} in the surface layer of the soil may be expected to reach the equilibrium value of about 247.4 - 500.3 mc/km^2.

Figs.13 - 16 show the ranges of the fallout rates of Sr^{90} and Cs^{137} at the Pacific side and at the Japan Sea side of the Japan mainland after the end of 1958 as estimated from the equation (26) assuming t=0 at the end of 1958 or at the beginning of 1959. (Fig.13) (Fig.14) (Fig.15) (Fig.16)

The simbols representing the different estimates in Figs.13 and 14 have the same meanings as those in Figs.1 and 2. As can be seen in these figures, as a general tendency the fallout rates of Sr^{90} and Cs^{137} may be expected to decrease theoretically after the end of 1958 when the nuclear testings were suspended by the three major powers possessing the nuclear weapons. However, because of the unusually high fallout rate in early 1959, the ranges of the fallout rates $F_{r(0)}$ at the end of 1958 had to be taken somewhat wider in these theoretical analyses for the future than those in the previous analyses.

Figs.17 - 20 show the ranges of the cumulative depositions of Sr^{90} and Cs^{137} at the Pacific side and at the Japan Sea side of the Japan mainland. After the end of 1958 as estimated from the equation (27) assuming t=0 at the end of 1958 or at the beginning of 1959. The symbols representing the different estimates in Figs.17 and 18 have the same meanings as those in Figs.5 and 6. (Fig.17) (Fig.18) (Fig.19) (Fig.20)

Because of the repeated nuclear testings up to the fall of 1958, the stratospheric fallout may continue for the future. Assuming the half elimination constant T_s from the stratospheric reservoir to be about two years, a maximum cumulative deposition may be observed at about five to six years after the end of 1958, as can be seen in these figures. At the Pacific side the range of the maximum cumulative deposition of Sr^{90} to be expected in 1963 - 1964 may

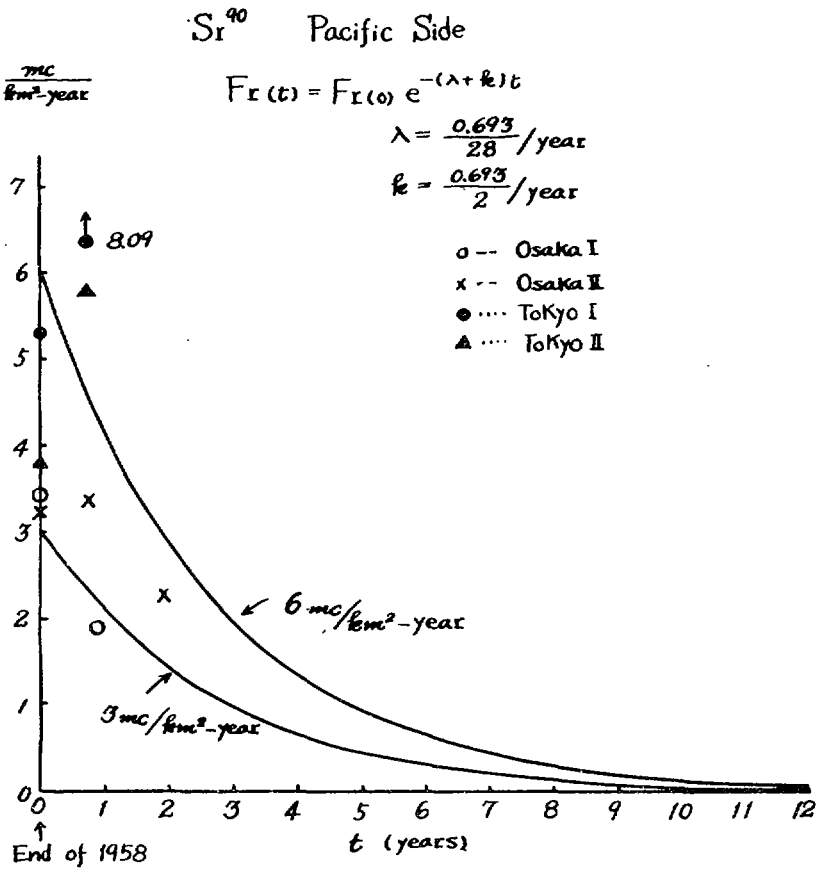

Sr^{90} Pacific Side
$\frac{mc}{km^2\text{-year}}$
$F_{r(t)} = F_{r(0)}\, e^{-(\lambda+k)t}$
$\lambda = \frac{0.693}{28}$/year
$k = \frac{0.693}{2}$/year
o -- Osaka I
x -- Osaka II
● ···· Tokyo I
▲ ···· Tokyo II
8.09
6 mc/km²-year
3 mc/km²-year
End of 1958
t (years)
0 1 2 3 4 5 6 7 8 9 10 11 12
0 1 2 3 4 5 6 7

Fig. 13

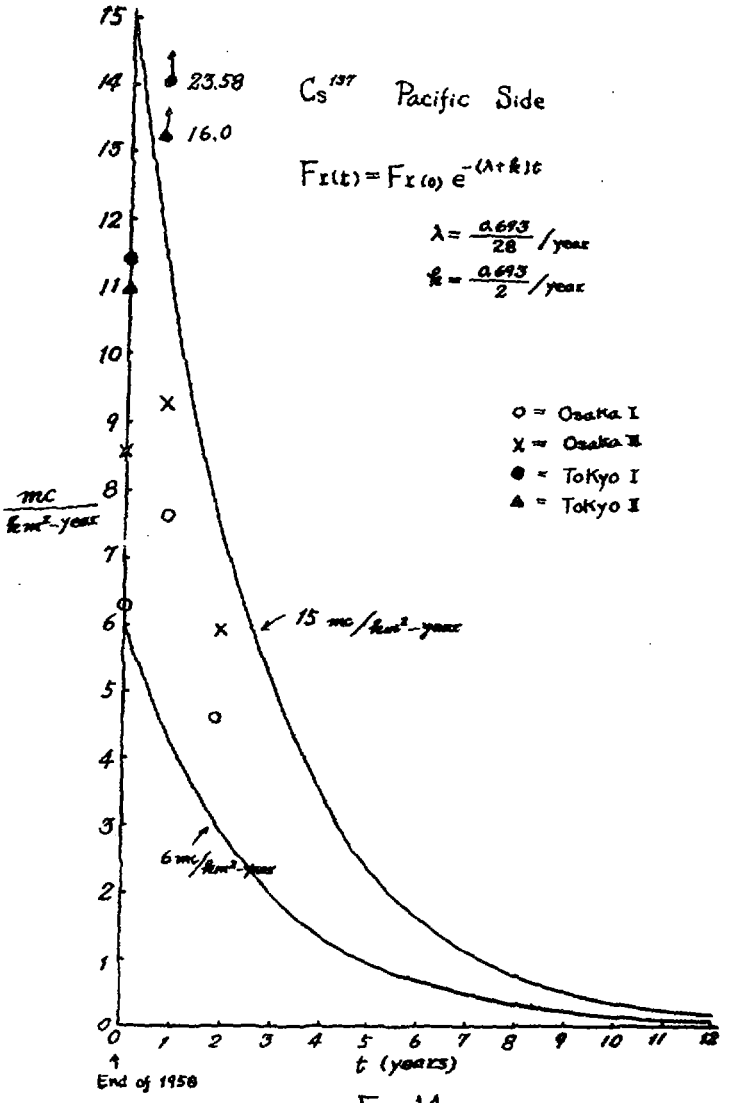

23.58
16.0
Cs^{137} Pacific Side
$F_{x(t)} = F_{x(0)} e^{-(\lambda+k)t}$
$\lambda = \frac{0.693}{28}$ /year
$k = \frac{0.693}{2}$ /year
○ = Osaka I
× = Osaka II
● = Tokyo I
▲ = Tokyo II
mc / ft m² - year
15 mc/ft m² - year
6 mc/ft m² - year
t (years)
End of 1958

Fig. 14

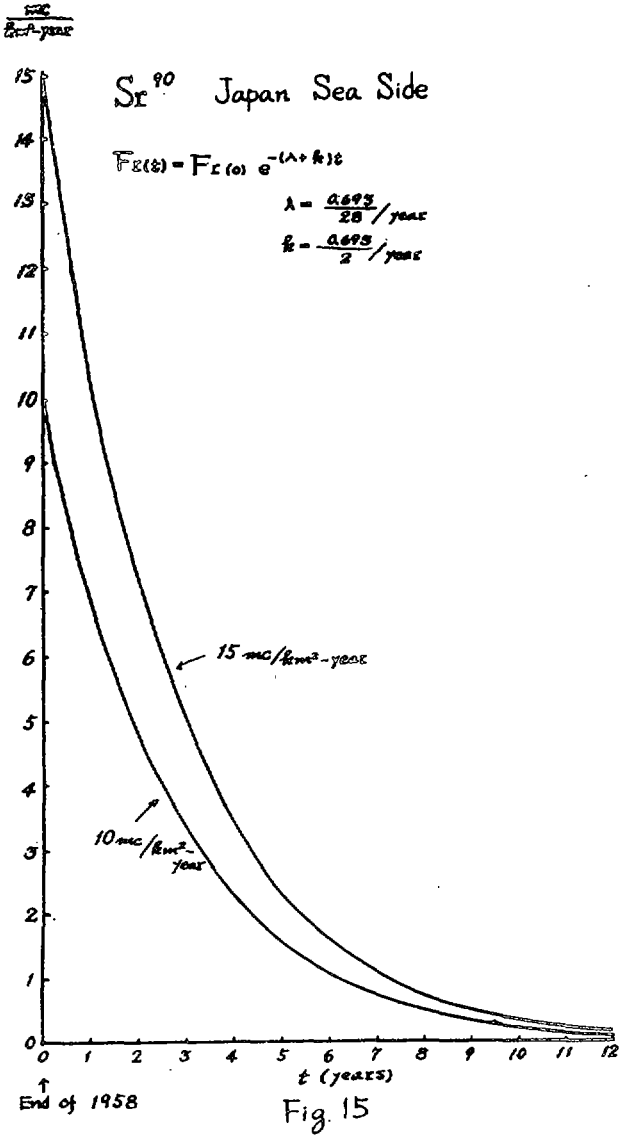

mc / km² - year
Sr 90 Japan Sea Side
$F_{\Sigma}(t) = F_{\Sigma}(0)\, e^{-(\lambda+k)t}$
$\lambda = \frac{0.693}{28}$ / year
$k = \frac{0.693}{2}$ / year
15 mc/km²-year
10 mc/km²-year
t (years)
End of 1958

Fig. 15

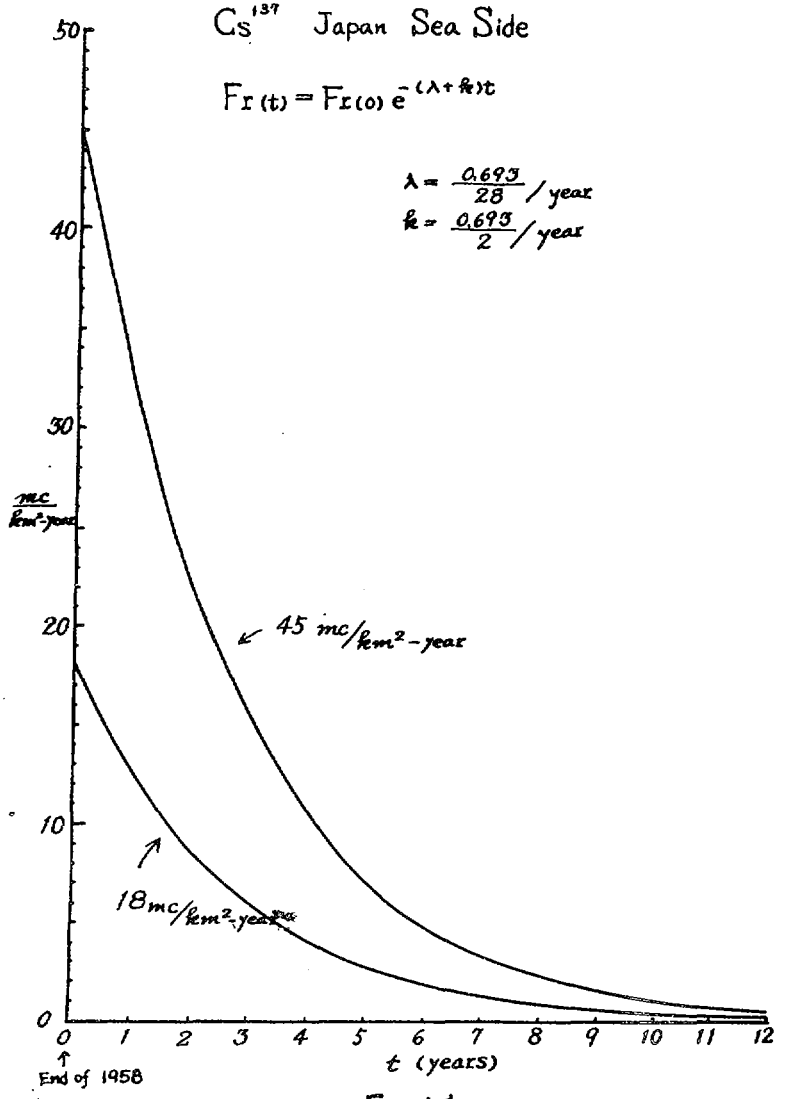

Cs 137 Japan Sea Side
Fr (t) = Fr (0) e^-(λ+k)t
λ = 0.693/28 / year
k = 0.693/2 / year
50
40
30
20
10
0
mc/km²-year
45 mc/km²-year
18mc/km²-year
0 1 2 3 4 5 6 7 8 9 10 11 12
t (years)
End of 1958

Fig. 16

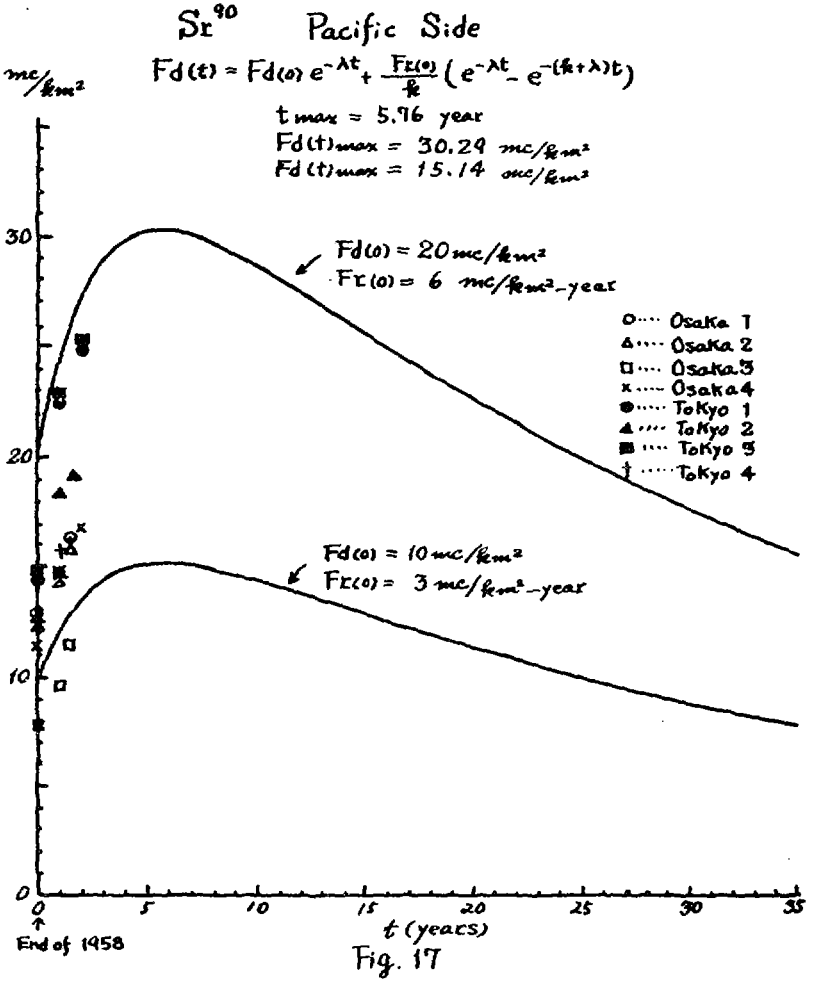

Sr 90 Pacific Side
Fd(t) = Fd(0) e^-λt + Fr(0)/k (e^-λt - e^-(k+λ)t)
mc/km²
t max = 5.96 year
Fd(t) max = 30.29 mc/km²
Fd(t) max = 15.14 mc/km²
Fd(0) = 20 mc/km²
Fr(0) = 6 mc/km²-year
Fd(0) = 10 mc/km²
Fr(0) = 3 mc/km²-year
Osaka 1
Osaka 2
Osaka 3
Osaka 4
Tokyo 1
Tokyo 2
Tokyo 3
Tokyo 4
30
20
10
0
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 17

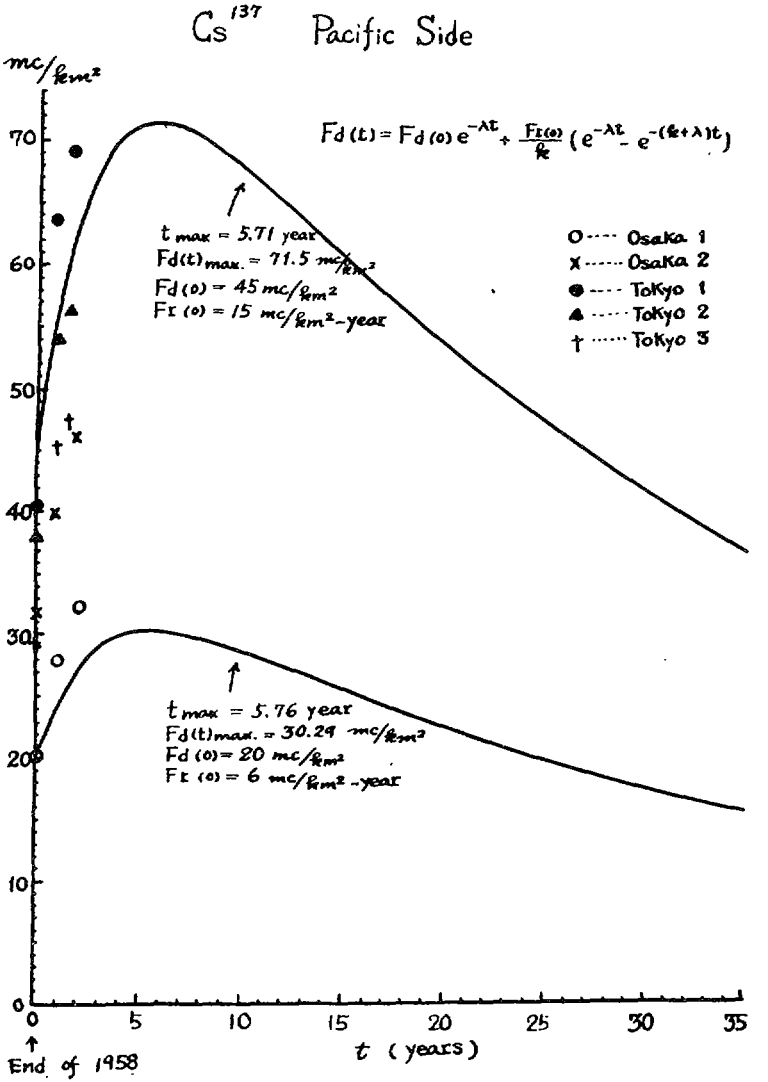

Cs 137 Pacific Side
mc/km²
Fd(t) = Fd(0) e^{-λt} + Fr(0)/k (e^{-λt} - e^{-(k+λ)t})
t max = 5.71 year
Fd(t) max. = 71.5 mc/km²
Fd(0) = 45 mc/km²
Fr(0) = 15 mc/km²-year
O ---- Osaka 1
x ------ Osaka 2
● ---- Tokyo 1
▲ ------ Tokyo 2
† ------ Tokyo 3
t max = 5.76 year
Fd(t) max. = 30.29 mc/km²
Fd(0) = 20 mc/km²
Fr(0) = 6 mc/km²-year
70
60
50
40
30
20
10
0
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 18

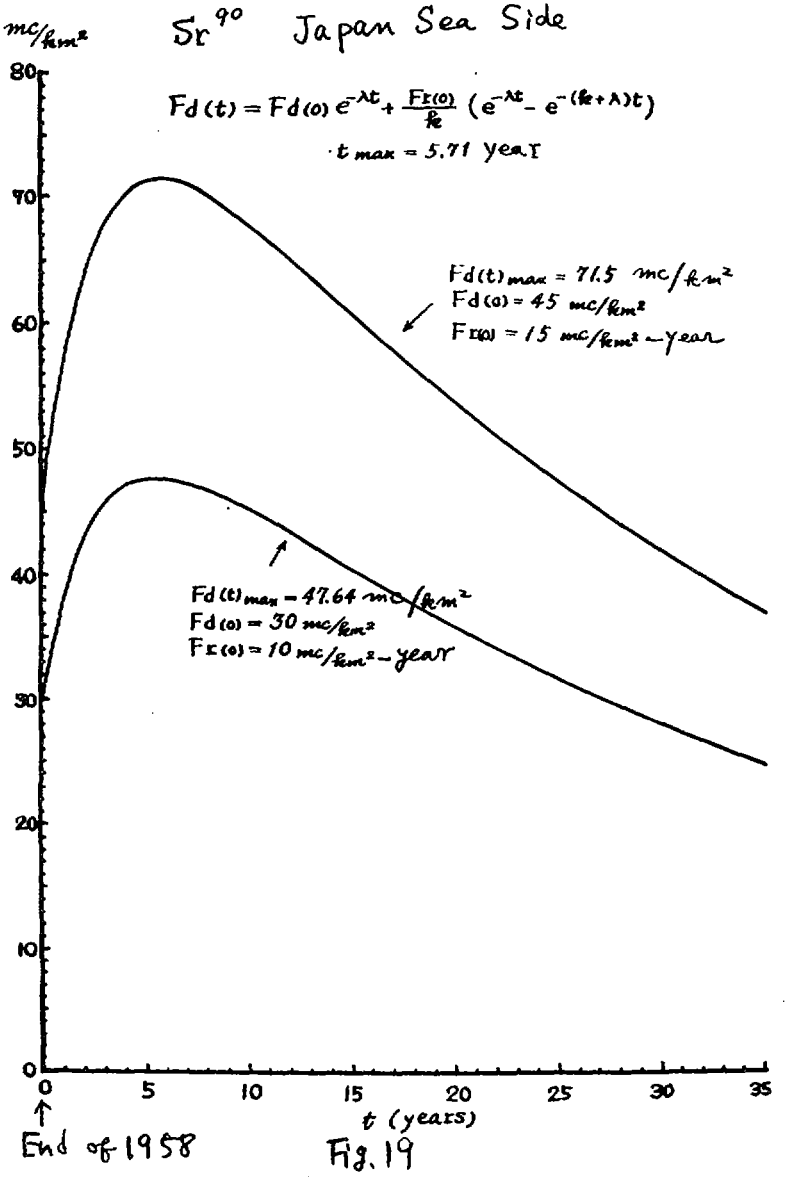

mc/km²
Sr⁹⁰ Japan Sea Side
$Fd(t) = Fd(0)\,e^{-\lambda t} + \frac{Fr(0)}{k}\,(e^{-\lambda t} - e^{-(k+\lambda)t})$
t max = 5.71 year
Fd(t) max = 71.5 mc/km²
Fd(0) = 45 mc/km²
Fr(0) = 15 mc/km² - year
Fd(t) max = 47.64 mc/km²
Fd(0) = 30 mc/km²
Fr(0) = 10 mc/km² - year
80
70
60
50
40
30
20
10
0
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 19

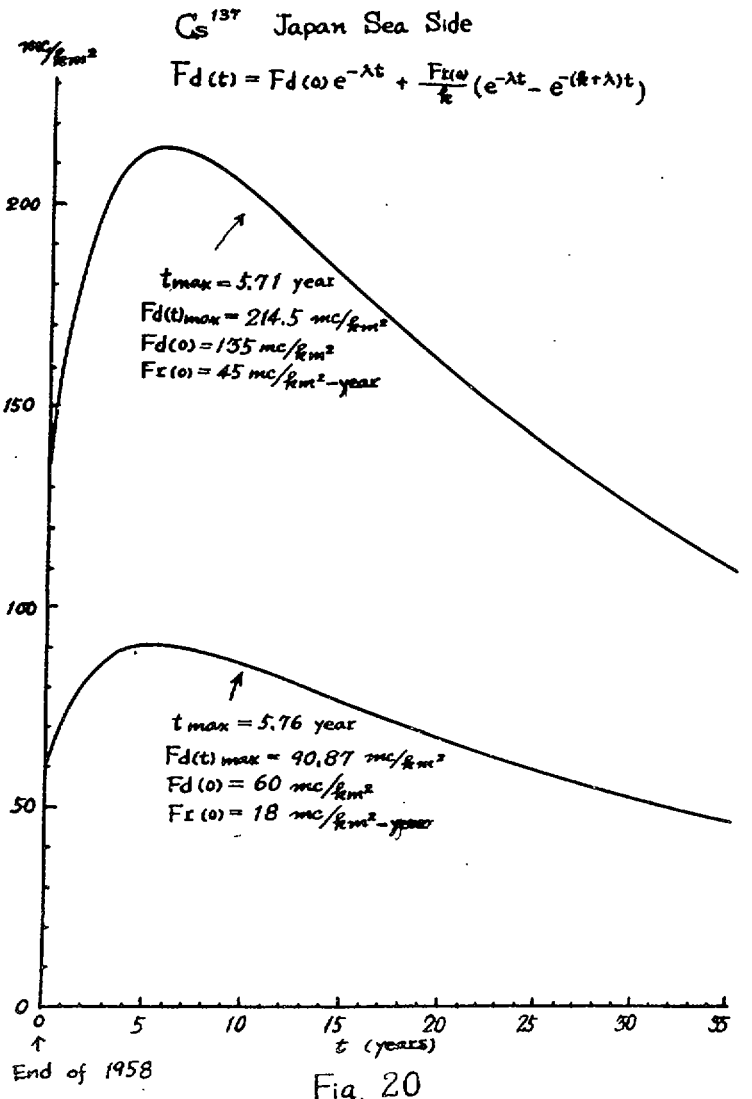

Cs 137 Japan Sea Side
Fd(t) = Fd(o) e^-λt + Fr(o)/k (e^-λt - e^-(k+λ)t)
mc/km²
200
150
100
50
0
tmax = 5.71 year
Fd(t)max = 214.5 mc/km²
Fd(o) = 135 mc/km²
Fr(o) = 45 mc/km²-year
t max = 5.76 year
Fd(t) max = 90.87 mc/km²
Fd(o) = 60 mc/km²
Fr(o) = 18 mc/km²-year
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 20

be about 15 - 30 mc/km^2 while that of Cs^{137} about 30 -72 mc/km^2. At the Japan Sea side the range of the maximum cumulative deposition of Sr^{90} to be expected in 1963 - 1964 may be about 47 - 72 mc/km^2 while that of Cs^{137} about 90 - 215 mc/km^2.

Figs.21 - 24 show the ranges of the ground accumulations of Sr^{90} and Cs^{137} in the surface layer of the soil at the Pacific side and the Japan Sea side of the Japan mainland after the end of 1958 as estimated from the equation (28) assuming t=0 at the end of 1958 or at the beginning of 1959 and the half weathering constant T_g to be about ten years. (Fig.21) (Fig.22) (Fig.23) (Fig.24)

As clear from the comparison of Figs.21 - 24 with Figs.17 - 20, if we assume a weathering factor of T_g=10 years, the ground accumulation in the surface layer of the soil may be expected to reach a maximum in two to three years after the end of 1958, much earlier than the total deposition without weathering effects. As can be seen in these figures, at the Pacific side of the Japan mainland the range of the maximum ground accumulation of Sr^{90} in the surface layer of the soil to be expected in the middle of 1961 may be about 12 - 24 mc/km^2 while that of Cs^{137} about 24 - 57 mc/km^2. From the preliminary analysis with the 6N HCl extraction of the soil about 10 cm from the surface sampled at the play ground of the Tokyo Institute of Technology, the ground deposition of Sr^{90} in the surface layer of the soil was estimated to be about 17 mc/km^2 at the end of August 1961.

Since this value is in the range of the ground accumulation of Sr^{90} in the surface layer of the soil at the Pacific side of the Japan mainland, the half weathering time T_g of about 10 years assumed in the above does not seem to be a figure too unreasonably misplaced, as far as the above result in Tokyo is concerned.

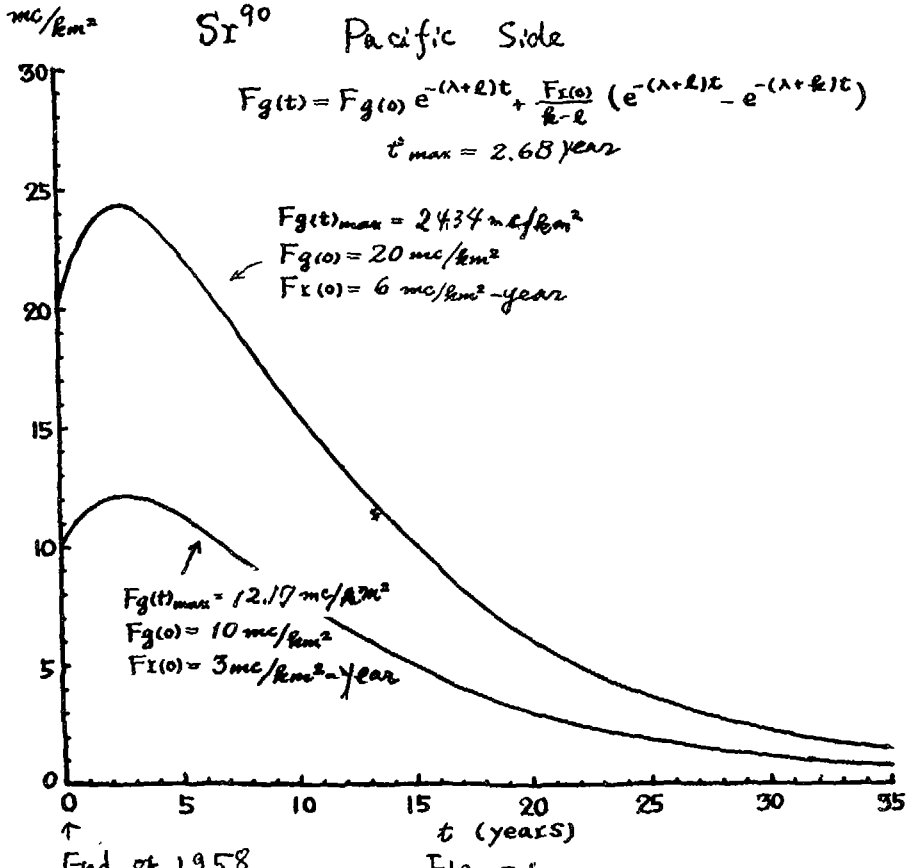

mc/km²
Sr⁹⁰ Pacific Side
$F_{g(t)} = F_{g(0)} e^{-(\lambda+\ell)t} + \frac{F_{x(0)}}{k-\ell}\left(e^{-(\lambda+\ell)t} - e^{-(\lambda+k)t}\right)$
$t'_{max} = 2.68$ years
$F_{g(t)max} = 24.34$ mc/km²
$F_{g(0)} = 20$ mc/km²
$F_{x(0)} = 6$ mc/km²-year
$F_{g(t)max} = 12.17$ mc/km²
$F_{g(0)} = 10$ mc/km²
$F_{x(0)} = 3$ mc/km²-year
30
25
20
15
10
5
0
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 21

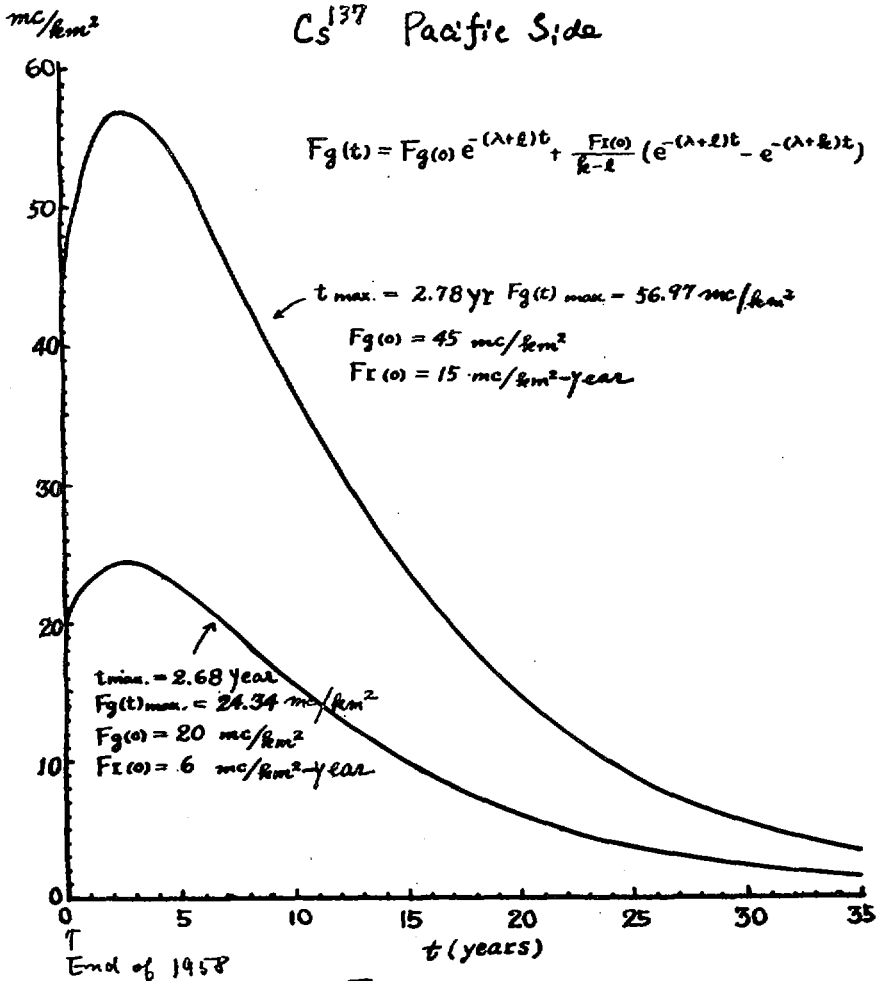

mc/km²
Cs¹³⁷ Pacific Side
60
50
40
30
20
10
0
0
5
10
15
20
25
30
35
t (years)
End of 1958
$F_g(t) = F_{g(0)} e^{-(\lambda+\ell)t} + \frac{F_{I(0)}}{k-\ell}(e^{-(\lambda+\ell)t} - e^{-(\lambda+k)t})$
t max. = 2.78 yr Fg(t) max. = 56.97 mc/km²
Fg(0) = 45 mc/km²
FI(0) = 15 mc/km²-year
t max. = 2.68 year
Fg(t) max. = 24.34 mc/km²
Fg(0) = 20 mc/km²
FI(0) = 6 mc/km²-year

Fig. 22

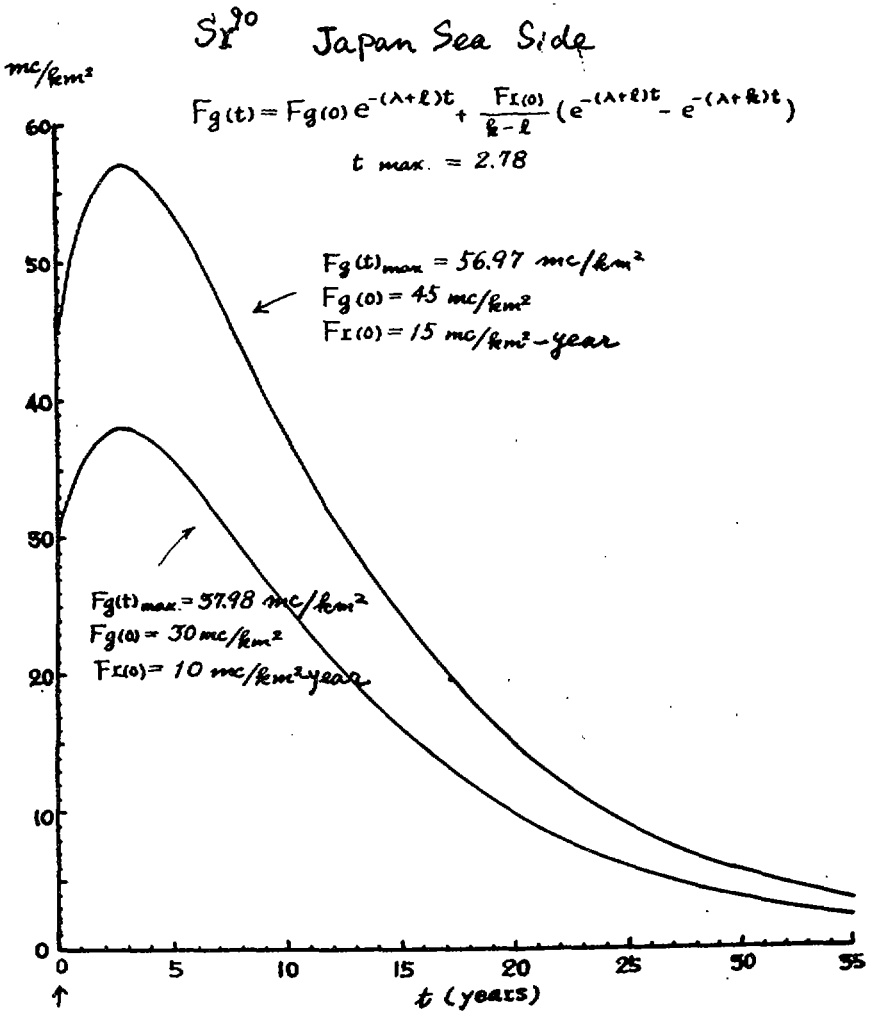

Sr^{90} Japan Sea Side
mc/km²
$Fg(t) = Fg(0)\, e^{-(\lambda+\ell)t} + \frac{FI(0)}{k-\ell}\left(e^{-(\lambda+\ell)t} - e^{-(\lambda+k)t}\right)$
t max. = 2.78
$Fg(t)_{max}$ = 56.97 mc/km²
Fg(0) = 45 mc/km²
FI(0) = 15 mc/km²-year
$Fg(t)_{max}$ = 37.98 mc/km²
Fg(0) = 30 mc/km²
FI(0) = 10 mc/km²year
60
50
40
30
20
10
0
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 23

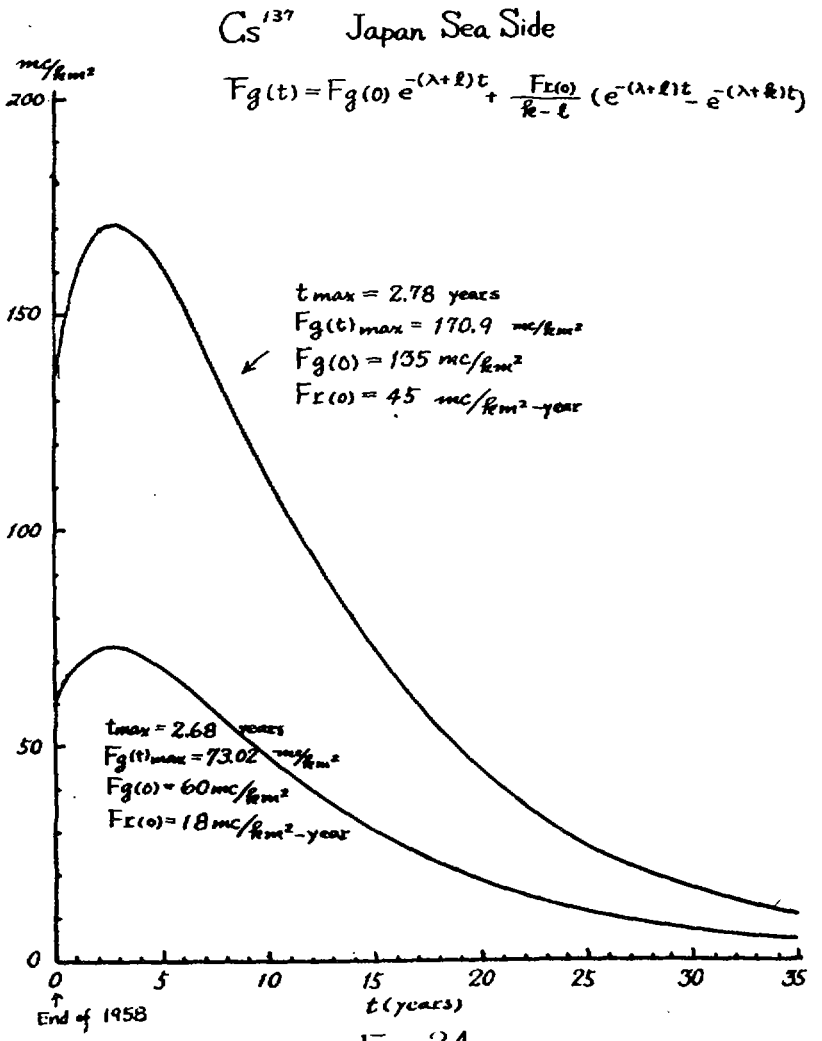

Cs137 Japan Sea Side
Fg(t) = Fg(0) e^-(λ+ℓ)t + Fr(0)/(k-ℓ) (e^-(λ+ℓ)t - e^-(λ+k)t)
mc/km²
200
150
100
50
0
tmax = 2.78 years
Fg(t)max = 170.9 mc/km²
Fg(0) = 135 mc/km²
Fr(0) = 45 mc/km²-year
tmax = 2.68 years
Fg(t)max = 73.02 mc/km²
Fg(0) = 60 mc/km²
Fr(0) = 18 mc/km²-year
0
5
10
15
20
25
30
35
t (years)
End of 1958

Fig. 24

However, the weathering effects may in general greatly depend on the various local conditions, especially the nature of the soil, the amount of the rainfall, and other various artificial factors such as irrigation, farming, ploughing, and the types and the extent of the grass or trees growing on the land, etc..

At the Japan Sea side the range of the maximum ground accumulation of Sr^{90} in the surface layer of the soil to be expected in the middle of 1961 may be estimated at about 38 - 57 mc/km^2 while that of Cs^{137} at about 73 - 171 mc/km^2.

(III) Dose Estimations due to Sr^{90} and Cs^{137}

(1) External irradiation

The external dose rate from the uniform diposition of gamma emitter on an infinite plane may be assumed to be proportional to the cumulative ground deposition $F_{r(t)}$ and the average energy E_γ of the gamma rays emitted from the deposit.

$$\left[\frac{dD}{dt}\right]_{ex} = k E_\gamma F_\gamma(t) \tag{37}$$

where the proportionality constant k may be assumed to be approximately k=0.1 mrad or mrem/year per one mc/km^2 and per one MeV of gamma ray energy. In case of Cs^{137}, since 92% of the disintegrations give gamma rays of energy 0.661 Mev and of these 11% are converted, the average gamma energy per disintegration may be estimated to be about E_γ=0.54 MeV. Substituting the above values into equation (37), we may obtain

$$\left[\frac{dD}{dt}\right]_{ex} = 0.055 \times F_\gamma(t) \text{ mrem/year} \tag{38}$$

where $F_{r(t)}$ is the cumulative ground deposition of gamma emitter in unit of mc/km^2. However, if we assume a shielding factor of about 5.5 for the living conditions of the average Japanese in the rural area, the above equation may be written as

$$\left[\frac{dD}{dt}\right]_{ex} = g_{ex} F_\gamma(t) \text{ mrem/year} \tag{39}$$

where

$$g_{ex}(Cs^{137}) \doteqdot 0.01 \frac{\text{mrem}}{\text{year}} \Big/ \frac{\text{mc}}{\text{km}^2} \tag{40}$$

(2) Internal irradiation

If we assume that the internal irradiation due to Sr^{90} and Cs^{137} would depend on both the fallout rate $F_{r(t)}$ and the cumulative deposition $F_{c(t)}$, the annual dose rate due to internal irradiation may be expressed in general by the following equation.

$$\left[\frac{dD}{dt}\right]_{in} = g_{i1} F_{r(t)} + g_{i2} F_{c(t)} \qquad (41)$$

where g_{i1} and g_{i2} are the proportionality constants.

In case of Sr^{90}, assuming about 0.5 mμc in the total bone of about 1 kg per mc/km^2 - year and about 0.15 mμc per mc/km^2 - year and the annual dose rate of about 15 mrem/year corresponding to 1 S.U. or a total burden of about 1 mμc in the bone of about 1 kg, we may have the following values for the constants g_{i1} and g_{i2} in equation (41)

$$g_{i1}(Sr^{90}) = 7.5 \frac{mrem}{year} \Big/ \frac{mc}{km^2\text{-}year} \qquad (42)$$

$$g_{i2}(Sr^{90}) = 2.25 \frac{mrem}{year} \Big/ \frac{mc}{km^2} \qquad (43)$$

In case of Cs^{137}, assuming a continuous body burden of about 1 mμc in the whole body of about 70 kg per mc/km^2 - year and about 0.05 mμc per mc/km^2 and the annual dose rate of about 0.16 mrem/year corresponding to a continuous total body burden of about 1 mμc in the whole body of about 70 kg, we may have the following values for the constants g_{i1} and g_{i2} in equation (41).

$$g_{i1}(Cs^{137}) = 0.16 \frac{mrem}{year} \Big/ \frac{mc}{km^2\text{-}year} \qquad (44)$$

$$g_{i2}(Cs^{137}) = 0.008 \frac{mrem}{year} \Big/ \frac{mc}{km^2} \qquad (45)$$

(3) Integral Dose

In case of Cs^{137} we may have to take into consideration both the external and the internal irradiation. Therefore the integral dose for the period of t_1 years may be given by the following general equation.

$$D_{t_1}(Cs^{137}) = g_{ex}\int_0^{t_1} F_r(t)dt + g_{i1}\int_0^{t_1} F_r(t)dt + g_{i2}\int_0^{t_1} F_c(t)dt \qquad (46)$$

In case of Sr^{90}, only the internal irradiation may be considered important and the integral dose may be given by the following equation.

$$D_{t_1}(Sr^{90}) = g_{i1}\int_0^{t_1} F_r(t)dt + g_{i2}\int_0^{t_1} F_c(t)dt \qquad (47)$$

Assuming $F_{r(t)}$ of equation (26) the integral may be given by

$$\int_0^{t_1} F_r(t)dt = F_{r(0)} I_r \qquad (48)$$

where

$$I_r = \frac{1}{k+\lambda}\left[1 - e^{-(k+\lambda)t_1}\right] \qquad (49)$$

Assuming $F_{d(t)}$ of equation (27) without weathering effects for $F_{r(t)}$ and $F_{c(t)}$ in the equations (46) and (47), the integral of $F_{d(t)}$ may be given by

$$\int_0^{t_1} F_d(t)dt = F_{d(0)} I_{d_1} + F_{r(0)} I_{d_2} \qquad (50)$$

where

$$I_{d_1} = \frac{1}{\lambda}\left(1 - e^{-\lambda t_1}\right) \qquad (51)$$

and

$$I_{d_2} = \frac{1}{k}\left[\frac{1}{\lambda}\left(1 - e^{-\lambda t_1}\right) - \frac{1}{k+\lambda}\left(1 - e^{-(k+\lambda)t_1}\right)\right] \qquad (52)$$

If we assume $F_{g(t)}$ of equation (28) with weathering effects for $E_{r(t)}$ and $F_{c(t)}$ in the equations (46) and (47), the integral of $F_{g(t)}$ may be given by

$$\int_0^{t_1} F_g(t)\,dt = F_{g(0)} I_{g1} + F_{r(0)} I_{g2} \tag{53}$$

where

$$I_{g1} = \frac{1}{\lambda+\ell}\left(1-e^{-(\lambda+\ell)t_1}\right) \tag{54}$$

and

$$I_{g2} = \frac{1}{k-\ell}\left[\frac{1}{\lambda+\ell}\left(1-e^{-(\lambda+\ell)t_1}\right) - \frac{1}{\lambda+k}\left(1-e^{-(\lambda+k)t_1}\right)\right] \tag{55}$$

Usually the value of t_1 in the above equations is assumed to be 30 years for the mean generation time in the estimation of the genetic effects due to Cs^{137}, and 35 years for the average life expectancy and 70 years for the average life span in the estimation of the somatic effects due to Sr^{90}. Therefore, for the convenience of estimation, the values of the integral I_r, I_{d1}, I_{d2}, I_{g1} and I_{g2} have been computed for t_1=30, 35 and 70 years and are given in Table 1. ←(Table 1)

Assuming the upper limits of Sr^{90} and Cs^{137} at the Pacific side of the Japan mainland as shown in Figs.17 and 18 or in Figs.21 and 22 to be the representave values for the Japanese population, the average integral doses due to Sr^{90} and Cs^{137} the children born at the end of 1958 in Japan might additionally receive because of the repeated nuclear testings in the past may be estimated easily using the values in Table 1 and are listed in Table 2.

(Table 2)

Table 1

t_1 (years)	Ir	I_{d1}	I_{d2}	I_{g1}	I_{g2}
30	2.69	21.1	53	10.54	28.3
35	2.69	23.4	59.7	10.59	28.5
70	2.69	33.2	88	10.6	28.53

Table 2 ; Integral dose without weathering effects (D) and with weathering effects (G)

a ; Sr^{90}

t_1 (years)	D (mrem)	G (mrem)
30	1786 (357)*	1310 (262)*
35	1979 (396)	1401 (280)
70	2803 (561)	1785 (357)

* The number in parenthesis indicates mrad with non-uniformity factor n=1.

b ; Cs^{137} (whole body)

t_1 (years)	D (mrem)	G (mrem)
30	38 (116)*	25 (83)*
35	41 (129)	27 (89)
70	57 (184)	35 (116)

* The number in parenthesis is the estimates without a shielding factor of 5.5 for external irradiation.

If we take into consideration the possible contribution of other bone seekers of shorter half-lives such as Sr^{89}, the total doses to the bone may be even higher than those estimated in Table 2. However, the average life-time dose to the bone marrow for the population would be somewhat less. Although the exact estimation of the average dose to the bone marrow would be rather difficult because of the various uncertainties involved in this type of estimation, judging from the Sr^{90} dose to the bone estimated in Table 2, the average total mean dose to the bone marrow for the population living up to age of 70 may be estimated roughly at about 1 - 3 rems.

In case of Cs^{137} because of its shorter biological half life or higher turnover rate as compared with Sr^{90} in the bone, the difference in the concentration between adults and children would not be as large as Sr^{90}. If we take into consideration the possible effects due to other fission products, the total doses to the whole body would be even higher than the double of those estimated only for Cs^{137} in Table 2, depending on the degree of contribution of other nuclides of shorter half-lives.

The 30 year dose from the external irradiation due to the cumulative deposition of grossactivity has been estimated at about 109 - 218 mrad without taking into consideration any shielding effects. Besides this there would be some contributions from the internal irradiation due to other fission products.

Therefore, taking these various factors into consideration, the total 30 year dose to the gonad may be estimated roughly at about 0.1 - 0.3 rems, or roughly about one tenth of the average life-time bone marrow dose.

(IV) Discussion and Summary

In this paper, based upon the theoretical considerations on the radioactive fallout due to nuclear testings prior to the end of August 1961, theoretical analyses on the fallout rate, the cumulative fallout deposition and the ground accumulation in the surface layer of the soil of Sr^{90} and Cs^{137} are attempted for the Pacific side and for the Japan Sea side of the Japan mainland. Based upon the theoretical analyses of the results up to the end of 1958, the possible future level of Sr^{90} and Cs^{137} is predicted assuming the half elimination time from the stratospheric reservoir to be about 2 years. In the estimation of the ground accumulation in the surface layer of the soil, the weathering effect with the half weathring time of about 10 years is assumed.

However, the result of the theoretical estimation with this assumption does not seem to be contradictory to the perliminary result of the radiochemical analysis of the surface layer of the soil up to a depth of about 10 cm conducted at the Tokyo Institute of Technology in 1961.

Because of a large amount of Sr^{90} and Cs^{137} injected into the stratosphere by the repeated large scale nuclear testings up to the end of 1958, the stratospheric fallout may continue for the future. Although the fallout rate may decrease with time, the total cumulative deposition may be expected to reach a maximum in about 5 - 6 years after the end of 1958 while the ground accumulation in the surface layer of the soil with the assumption of half weathering time of about 10 years in about 2 - 3 years. These results are summarized in Table 3. Although the fallout rate may be somewhat higher at the Japan Sea side than at the Pacific side of the Japan mainland, assuming the upper limit for the Pacific side as the representative value for the Japanese population because of the larger percentage of population living along the Pacific side, the possible integral doses to the bone due to Sr^{90} and to the whole body due to Cs^{137} were estimated as shown in Table 2.

(Table 3)

Table 3 ; Maximum cumulative deposition without weathering effect and maximum ground accumulation in the surface layer of the soil with weathering effect to be expected at t_{max} years after the end of 1958

Site / Maximum deposition at t_{max}		Pacific Side		Japn Sea Side	
		lower limit	upper limit	lower limit	upper limit
Sr^{90}	$Fd(t)_{max}$ (mc/km²)	15	30	48	72
	t_{max} (year)	5.8	5.8	5.7	5.7
	$Fg(t)_{max}$ (mc/km²)	12	24	38	57
	t_{max} (year)	2.7	2.7	2.8	2.8
Cs^{137}	$Fd(t)_{max}$ (mc/km²)	30	72	91	215
	t_{max} (year)	5.8	5.7	5.8	5.7
	$Fg(t)_{max}$ (mc/km²)	24	57	73	171
	t_{max} (year)	2.7	2.8	2.7	2.8

However, taking into consideration the possible contribution from other fission products, the total life time bone marrow dose may be estimated roughly at about 1 - 3 rems while the 30 year dose to the gonad roughly at about 0.1 - 0.3 rems.

According to Lewis,[9] the probability for leukemia is estimated at about 2×10^{-6} per year per rem which may be valid within a factor of about 3, while UN report[2] assume the probability of about 1.5×10^{-6} per year per rem to continue after each element of radiation exposure for the remaining life of the individual.

Using these probabilities for leukemia and assuming the linear relationship between the dose and the effect, the total number of leukemia for a population of about 10^8 may be estimated to be about $5 - 7 \times 10^3$ corresponding to about 1 rem of the total mean marrow dose. However, if we take into consideration a factor of uncertainty of about 3 in the Lewis' probable estimate on the incidence of leukemia, the upper limit may be estimated at about 2×10^4. Although there may be various uncertainties in these estimates, judging from these values, the total number of such fatal somatic damage as the leukemia may be assumed to be roughly at about the order of magnitude of $0.5 - 1.0 \times 10^4$ corresponding to the total mean marrow dose of about one rem per person.

Therefore, if we assume a total mean marrow dose of about 3 rems, the total number of cases may be estimated about three times higher and roughly at about $1.5 - 3 \times 10^4$ for a toal population of about 10^8, as shown in Table 4 (a). ← (Table 4(a))

The genetic damage to the population may be estimated with the method as given by Crow[10] in the Fallout Hearings at the U.S. Congress.

Assuming a doubling dose of about 40 rems instead of 50 rems assumed by Crow and a normal incidence of tangible genetic defects due to spontaneous mutation to be about 2 - 4%,[2] the total number of gross physical or mental defects corresponding to a 30 year gonad dose of about 0.1 rem for a stable total population of about 10^8 may be estimated as follows.

Table 4 ; Numerical estimation of the possible damage for the population of about 10^8 in Japan due to the past nuclear testings conducted during the period from early 1954 to the end of August 1961

(a) ; Leukemia

	lower estimate	higher estimate
Leukemia	5,000 ~ 10,000	15,000 ~ 30,000

$$\frac{0.1}{40} \times \frac{2\text{-}4}{100} \times \frac{10^8}{2} = 2.5\text{-}5 \times 10^3 \qquad (56)$$

Assuming about 10% expression in the first generation, the first generation effect may be estimated at about 250 - 500. Assuming the frequency of stillbirths and childhood deaths at mutational equilibrium to be about 8% as estimated from increased death rates in children of consanguineous marriages,[15] the total number of stillbirths and childhood deaths from 0.1 rem fro a generation may be estimated.

$$\frac{0.1}{40} \times \frac{8}{100} \times \frac{10^8}{2} = 10^4 \qquad (57)$$

Assuming about 6% expression in the first generation, the first generation effect may be estimated at about 600.

Russell[16] observed a 3% reduction in litter size of mice at 3 weeks of age when the sires had been exposed to 300 r. Using this result and assuming that the total effect would be duubled when both parents were exposed, the first generation effect of the embryonic and neonatal deaths may be estimated.

$$\frac{2(0.1)}{300} \times \frac{3}{100} \times \frac{10^8}{2} = 10^3 \qquad (58)$$

Assuming this number of the first generation effect would correspond to about 6% of the total effect, the total damage may be estimated roughly at about 1.7×10^4.

If the 30 year gonad dose is assumed to be 0.3 rem, the genetic damage estimated in the above would be about three times higher than the above estimates. The results of these numerical estimations of the possible genetic damage due to the radiation effects of fission products for a population of about 10^8 are summarized in Table 4 (b). ← Table 4(b)

Although these estimated numbers may be considered subject to a large uncertainty, it should be remembered that those listed in the

Table 4 (b); Genetic damage due to Fission Products

Generation / Type of effect	1st Generation		Total to infinity	
	lower estimate	higher estimate	lower estimate	higher estimate
Gross physical or mental defects	250 ~ 500	750 ~1,500	2,500 ~5,000	7,500 ~15,000
Stillbirths and childhood deaths	~ 600	~1,800	~10,000	~30,000
Embryonic and neonatal deaths	~ 1,000	~3,000	~ 17,000	~50,000

table are only the tangible ones and that there would be perhaps a larger but unknown number of minor or intangible defects as pointed out by Dr. Crow[10] at the Fallout Hearings in the United States.

Besides the possible effects due to the radiation from fission products, we must take into consideration the possible effects of carbon-14 as has been repeatedly emphasized by Prof. Linus Pauling.[11] Leipunsky[12] and Totter, et al[13] have also made the estimation of the possible hazards due to the bomb produced carbon-14.

Assuming the probability[3] of any given generative cell having had a carbon-14 transmutation in DNA to be about 6×10^{-4}, for a population of 2.5×10^{9} with a birth rate of 30 per 1000 the total number of children with the transmutation effects in its genetic material may be estimated as follows.

$$(2.5\times10^{9})\times\left(\frac{30}{1000}\right)\times(6\times10^{-4}) \doteq 45\times10^{4} \qquad (59)$$

Leipunsky assumed in his estimates the ratio of mutations to transmutations (M/T) to be unity and an increment of carbon-14 due to 10 megatons of fusion equal to 4.1×10^{-3} times the present carbon-14 equilibrium value. With this assumption Totter, et al estimated that the total number of people with defective genes due to the carbon-14 increment may be estimated to be as follows.

$$(4.1\times10^{-3})\times(45\times10^{4})\times(8100) = 1.44\times10^{6} \qquad (60)$$

Totter, et al. pointed out that the amount of carbon-14 estimated by Leipunsky would be higher by a factor of about four as compared with the estimated by Libby,[14] and that the ratio M/T might be much smaller than unity partly because of the existence of inert "non-genic" DNA in the cell in which C^{14} decay is less apt to cause mutation and partly because of a larger possibility of total inactivation of the cell with the transmutation effects in its genetic material, although there might be some possibility that one transmutation may cause more than one mutation.

However, if we assume the total of past nuclear testings prior to the end of August 1961 to be roughly about 100 megaton fission and 100 megaton fusion, the total increment of carbon-14 may be estimated roughly at about 1 - 2% of the present natural carbon-14 equilibrium value. Assuming an increment of about 1% and an immediate mixing of the carbon-14, the annual dose rate due to bomb-produced carbon-14 may be estimated roughly at about 0.015 mrem/year. With this small increment of annual dose the first generation effect may be undetectable. However, because of its longer half life of about 5,600 years the small effects may continue many thousands of years and the total number of damage may not be completely negligible. The total number of damage due to radiation corresponding to an increase of carbon-14 by about 1% above the natural carbon-14 level was estimated for a stable total population of about 10^8 as in the case of the fission products and is shown as a lower estimate of the total damage due to carbon-14 in Table 4 (c). Besides the effects due to radiation, there would be some transmutation effects, if not very much larger than the corresponding radiation effects in case of carbon-14 for the mammalian cells as pointed out by Totter, et al. However, since the exact estimation is difficult without knowing the ratio M/T as discussed in the above, the transmutation effect was assumed to be about equal to the radiation effect and the estimate of the combined effects is shown in the table as a higher estimate. Therefore this higher estimate may be considered to correspond to the radiation effects for an increment of carbon-14 of about 2% above the natural carbon-14 level. In about 5,600 years following the addition of the carbon-14 increment roughly about one half of the total number of damage due to carbon-14 given in the table may be expected to occur, although the number per generation may be considered almost negligible for a population of about 10^8.

← (Table 4(c))

As can be seen from the numerical estimations of various possible damages in the table, although the probability per person would be very small, the absolute number of possible long term damage may not always be considered negligible and it should be pointed out that a small additional dose to a large number of people may, in some

Table 4 (c) ; C^{14} damage

Period / Type of effect	Total in 5,600 yrs		Total to infinity	
	lower estimate	higher estimate	lower estimate	higher estimate
Gross physical or Mental defects	1,500 ~3,000	3,000 ~6,000	3,000 ~6,000	6,000 ~12,000
Stillbirths and childhood deaths	~6,000	~12,000	~12,000	~24,000
Embryonic and neonatal deaths	~10,000	~20,000	~20,000	~40,000

cases, ultimately bring about a greater number of total damage to the population at large than a very large lethal dose to a very small limited number of people. When a small number of people are exposed to a lethal dose, they may die without bearing a child and in this case no genetic damage may be transmitted to the rest of the population. However, if a closed total population is exposed to a continuous low dose of radiation in a wholesale manner, the genetic burden of the whole population may be expected to increase gradually in the natural background level.[17][18]

Besides the leukemia and the genetic damage, we must also consider the possible life-shortening effect due to radiation. Although it is reported by Lorenz[22] that the radiation may have a life-extending effect on mice at very low dosage region, we may assume on the conservative side that the life-shortening effect of radiation obtained at higher dosage could be extrapolated to the low dosage region, because of the various posible uncertainties inherent in the analysis of the results at very low dosage region. Depending on whether we assume the dose to the whole body is more responsible for the general life-shortening, there would be some difference in the estimates. However, in view of the lack of more accurate knowledge about this point, we may assume that the life-time dose responsible for the life-shortening effect would be about the order of magnitude of one rem in total due to the repeated nuclear testings prior to the end of August 1961, and that there would be no threshold of radiation dose in causing the general shortening of life span. With this assumption, assuming the life-shortening constant or the chronic lethality coefficient to be about $1.7 - 3.5 \times 10^{-4}$ as deduced from the data given by Brues and Sacher,[19] the average loss of life span per person in number of

days may be estimated to be roughly about 4.3 - 9 days for a population with the average normal life span of about 70 years.

The average loss of life span, although it appears to be very small per person, and well within the limit of statistical error, may be considered to correspond to a total loss of life span equivalent to about 1.2 - 2.5 x 10^6 man-years for a population of about 10^8. If we assume the dose responsible for life-shortening to be about 3 rems, the resulting estimate would be about three times higher than the above value and may be estimated at about 3.6 - 7.5 x 10^6 man-years. Assuming a total loss of life span of about four million years which may correspond to about 14.6 days or roughly about two weeks per person on the average, it may be considered numerically to correspond to an average loss of life span of about 40 years of 100,000 people.(20)

There may probably be a large factor of uncertainty of about ten or more in the above numerical estimation of the possible hazards to man because of the various assumption introduced in the course of estimation. Moreover, whether there would be threshold or not for the long-term radiation effects considered in this paper is still a matter of controversy even among the specialists on the subjects, although the evidence seems to be accumulating to indicate that there would probably be no threshold in the genetic effect or that the threshold would be very low, if it existed. Therefore the numbers introduced in the above should be interpreted as such. However, because of such a large factor of various uncertaintes in this basic scientific data, on which the above estimations are based, we feel it side. From these standpoints, we must estimate the maximum possible dangers that could be conceivable within the limit of large uncertainties and try to exert ourselves to minimize the possible risks for the population.

Table 4 (d): Life shortening effects

	Average per person	Total loss for 10^8
Lower Estimate	4.3 ~ 9 days	1,200,000 ~2,500,000 man-years
Higher Estimate	12.9 ~ 27 days	3,600,000 ~7,500,000 man-years

Otherwise, if we took a too optimistic assumption inspite of a large factor of uncertainties and if it should prove in future that the situation would be pessimistic, it would be too late to do anything for it many years after the various injuries had taken place or had been detected or proven among the people. Therefore,

(Table 4(á))

it seems to the author only right in dealing with any matter concerning human life to take account of all possible dangers which may be conceivable in the light of our present scientific knowledge. In this respect, whether the above estimates would be ten times higher or ten times less becauses of the various uncertainties involved in the estimation is not very important. The point is that the possible number of people who would be seriously hurt in the future can not be considered absolutely zero because of a large factor of uncertainty about the long-term radiation effects. From the stand point of public health or conscientious medical science at peace time, in which every effort must be made to save even a single life and to make life even one day longer, even a slight "unnecessary" increase of radioactivity above the naturally occurring level, due to repeated nuclear testings is certainly undersirable.

From the peaceful uses of radiation and atomic energy, such as the medical diagnostics or treatments by radiation, the atomic power plant, or the industrial uses of radiation, we may get more benefits than the possible risks.

However, what benefit do we get from the repeated nuclear testings conducted by major powers preparing for a genocide of hundreds of millions of people? It is the problem of deep concern of those who are trying to develop peaceful uses of atomic energy for the peace, happiness and welfare of mankind, that the allowances of the permissible level of radiation and radioactivity would be gradually narrowed down with the gradual increase of the level of radioactive contamination of our environment due to large scale nuclear testings.(21)

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